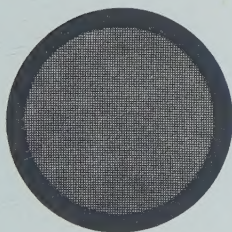
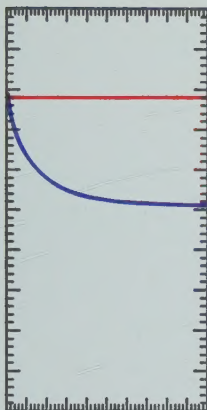


CHRISTER SAMUELSSON



^{222}Rn AND DECAY PRODUCTS IN OUTDOOR AND INDOOR ENVIRONMENTS;

Assessment Techniques Applied to Exhalation ,

Air-Cleaning and Arctic Air.

*Diss. Lund.
1984*

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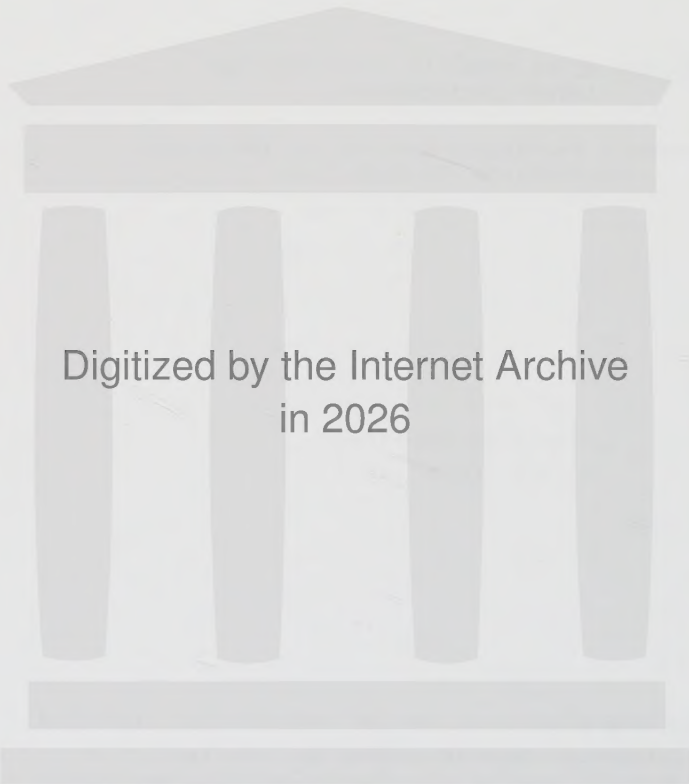
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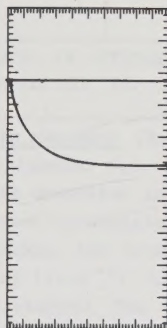
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Air-Cleaning and Arctic Air.

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Christer Samuelsson

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Title and subtitle ²²² Rn AND DECAY PRODUCTS IN OUTDOOR AND INDOOR ENVIRONMENTS; Assessment Techniques Applied to Exhalation, Air-Cleaning and Arctic Air.	Sponsoring organization	
	Abstract <u>Radon-222</u> (radon) and <u>radon daughter</u> (RnD) measurement methodologies are analysed from both theoretical and experimental points of view. It is shown that exhalation from enclosed porous materials can be described in terms of the <u>time-dependent diffusion</u> theory. Deficiencies in the established accumulator method of radon <u>exhalation</u> measurement are shown. By the existing methods, the true free exhalation rate of thin samples may be underestimated by a factor of $(1 + \alpha^{-1})$, in radon-tight accumulators (α is the outer to inner volume ratio of the sample). The term 'back-diffusion' is clarified and shown applicable to steady-state conditions only. The <u>wire-screen</u> technique is utilized to separate aerosol-attached and <u>unattached</u> RnD in a 3 m³ radon cell. The effect of <u>air-filtration</u> on the RnDs is expressed as individual activity concentrations as well as in terms of <u>effective dose equivalent</u> rate, $\dot{H}$. $\dot{H}$ has been reduced by a factor between 1.3 and 2.5 for the small-sized areosol particles used (surface area median less than 100 nm), at the filtration rate constant 5 h⁻¹. The exact reduction value is dependent on initial <u>aerosol</u> load, type of <u>filter</u>, and dose model (Jacobi-Eisfeld and James-Birchall in this investigation). The concentration of radon and Pb-210 in the <u>Arctic</u> summer air averaged 75 ± 21 and 0.075 ± 0.028 mBq m⁻³, during the Swedish <u>Ymer-80</u> expedition. It is shown that steady-state equilibrium models are unsuitable for estimation of the mean aerosol residence time in ocean air. A good qualitative agreement between radon-levels and the time since the air mass left larger land areas was found. The radon-222 and long-lived daughter (<u>Pb-210</u>, <u>Po-210</u>) measurements are insensitive to ship- and local contaminations.	
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Signature Christer Samuelsson

Date May 2, 1984

There're still songs to be sung.

to

Ingrid, Henrik, Martin and Jenny.

This thesis is based on the following papers, which in the text will be referred to by their Roman numerals:

- I. C. Samuelsson and H. Pettersson,
Exhalation of ^{222}Rn from Porous Materials.
International Seminar on Indoor Exposure to Natural Radiation
and Related Risk Assessment, October 3-5, 1983, Anacapri, Naples, Italy.
(To be published in a special issue of Radiation Protection Dosimetry).
- II. C. Samuelsson and H. Pettersson,
A Continuous True Radon Monitor.
Proc. 3rd Int. Symp. on Radiation Protection.
Advances in Theory and Practice, Inverness, Scotland,
June 6-11, 1982.
- III. C. Samuelsson,
The Effect of Air Filtration on Aerosol-Attached and Unattached
 ^{222}Rn Decay Products.
Report LUNDF6/(NFRA-3041)/(1-38)/(1983)
- IV. C. Samuelsson, L. Hallstadius, B. Persson, R. Hedvall,
E. Holm and B. Forkman,
 ^{222}Rn and ^{210}Pb in the Arctic Summer Air;
Assessment Techniques and Results from the Swedish Expedition Ymer-80.
(Submitted for publication in Journal of Environmental Radioactivity).

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1. INTRODUCTION

In the last decade or so the interest in radon and radon decay products in the human environment has grown considerably. The reason is mainly epidemiological, and naturally the enhanced awareness of risks in the modern society is a trend, also supporting the radon issue.

There are three key factors, which have influenced our concern about the health effects of radon irradiation:

- i. The establishment of a positive and significant correlation between lung cancer incidence rate and radon daughter exposure in mines, during the 1960s;
- ii. The large-scale tendency towards energy saving in houses, by lowering the air exchange rate, thereby increasing the radon levels;
- iii. The introduction of a weighting procedure by the International Commission on Radiological Protection (ICRP77a) makes it possible to compare the risk of an inhomogeneous lung irradiation with that of an external whole body irradiation.

Besides the health aspects of radon and its decay products, there are, or have been, several applications of radon; in meteorology, geology, water research and medicine. In this study paper IV deals with radon and its decay products as natural trace elements in the atmosphere.

Before proceeding to a more definite listing of the objectives of the present investigation let us look first at the natural radioactive decay series, thereby giving a more precise meaning to 'radon' and 'radon daughters' used loosely above. This will be followed by a short historical review.

1.1. RADON IN THE NATURAL DECAY SERIES

In our radiation environment isotopes of radon and their decay products have always played an important role. All three naturally occurring decay series involve a radon variant. (The neptunium $4n + 1$ series, of zero natural abundance, does not include any radon isotopes!). In the ^{238}U series the radon isotope has mass number 222, and follows immediately after ^{226}Ra .

The radon members of the thorium and actinium series are ^{220}Rn ($T_{1/2} = 55.6 \text{ s}$) and ^{219}Rn ($T_{1/2} = 3.96 \text{ s}$) respectively. Mainly due to their short half-lives, these two radionuclides are of less significance in most circumstances, especially ^{219}Rn .

The present investigation is exclusively devoted to ^{222}Rn and its decay products. ^{222}Rn will in the following text alternatively be termed 'radon', where there is no possibility of confusion with the other radon isotopes.

The radionuclides of the ^{238}U series are shown in Figure 1 in paper IV. The progenies following radon in the series are popularly called 'radon daughters', and they can be divided into the short-lived group: ^{218}Po , ^{214}Pb , ^{214}Bi , ^{214}Po ; and the long-lived group ^{210}Pb , ^{210}Bi , ^{210}Po . In the following we will frequently designate the short-lived group by RnD , not to be confused with the old historical name for ^{210}Pb , RaD .

1.2. RADON AND RADON DAUGHTERS - A HISTORICAL REVIEW

Our knowledge of radon and its decay products is nearly as old as the history of radioactivity. Only a few years after Becquerel's discovery of radiation from uranium ores in 1896 Rutherford writes: 'I have found that thorium compounds continuously emit radio-active particles of some kind, which retain their radio-active powers for several minutes.' (Ru00). The emission of particles was termed 'emanation', and it was soon discovered that also radium and actinium had their own characteristic emanations. The suggestion by Rutherford that 'the emanation may be a vapour' (Ru00), was soon confirmed. The radium emanation was 'a new gas possessing the property of radioactivity to a very intense degree' (So09). In addition to the basic issue of how the radioactive elements could transform into each other, e.g. uranium into thorium (isotopy was unknown until 1913), the behaviour of emanation was puzzling. '...The emanation possesses a very remarkable property... of producing radio-activity in all substances on which it falls.' (Ru00), 'I have shown that radium emanation coheres to almost everything with which they come into contact', 'the persistence of radio-activity on glass vessels which have contained radium is remarkable. Filters, beakers, dishes used in the laboratory for operations with radium, after having been washed in the usual way, remain radio-active;' (Cr03). These early experiences of the tendency of radon daughters to adhere to surfaces, gave them their old name 'active deposit'. The individual members of the 'active deposit of rapid change' were soon discovered and given the names RaA , RaB , RaC and RaC' (Figure 1 paper IV). Of all the natural radionuclides, except for ^{238}U , it was actually

a long-live daughter, ^{210}Po (RaF), to be discovered first by the Curies in 1898. By 1908 all major radionuclides in Figure 1 IV were individually known.

The history of radon has also epidemiological and medical sides. That miners often got a lung disease, called 'Bergkrankheit' in German, had been known since the sixteenth century. The lung cancer incidence among miners was about 50 times greater than normal, during the decades around 1900 in Central Europe, (Ar67, Ed83). The 'Bergkrankheit' was later identified as lung cancer.

In the early decades of this century radon was used for intracavitary tumour radiation instead of radium. In Sweden there was no significant change-over from radium to radon (Si50), but abroad, mainly in the U.S.A., radon 'cows' were common during the twenties and thirties. Another, more or less historic application of radon, is as a healing agent in air and water of certain spas. Astonishingly the first tracer experiment was claimed to have taken place in a human food stuff. To prove to his denying landlady that the rest of the Sunday meal was served again, Hevesy conducted an experiment: 'The coming Sunday in an unguarded moment I added some active deposit to the freshly prepared pie and on the following Wednesday, with the aid of an electroscope I demonstrated to the landlady the presence of the active material in the soufflé.' (Gl67). Later Hevesy used thorium (^{212}Pb $T_{1/2}=10.6$ h) to indicate the uptake of lead in the horsebean (Vicita Faba), and this was the first published radioactive tracer application in biology (He23).

The presence of natural radioactivity in the atmosphere was proved for the first time 1901 by Elster and Geitel and soon quantitative measurements were performed. As early as 1917 Schweidler correlated high radon concentration in outdoor air with decreasing atmospheric pressure on warm days. Early references to the field of radioactivity in geophysics are found in the comprehensive book by Meyer and Schweidler (Me27). As meteorological parameters affected the distribution of radon and RnD in the atmosphere, it soon became clear that these nuclides could be used as natural tracers in outdoor air. For example, the vertical mixing coefficient due to air turbulence could be determined from measurements of the radon concentration height profile. References to the modern literature covering the subject of 'radon meteorology' can be found in the review papers by Schumann (Sc72) and Wilkening (Wi81).

1.3. THE AIM OF THE INVESTIGATION

In spite of more than 80 years experience in radioactivity, there still remain several unsolved problems concerning radon and radon daughters. In such basic areas as radon diffusive transport and radon daughter behaviour in indoor air, our knowledge is far from reliable. In the field of meteorology, the role of radon and radon daughters in atmospheric processes is poorly understood, even today. In general terms the aim of this investigation is to tackle unsolved radon problems. There are too many to be treated by one research group or laboratory, and naturally we have to limit the investigations to a few well defined areas.

Specifically the objectives of this study are:

1. to examine radon exhalation from porous media;
2. to evaluate the effect of air filtration on the radon daughter concentration in indoor air;
3. to apply radon- and radon daughter measurement methodologies to low outdoor activity levels.

Objectives 1 and 2 are directly associated with the health effects of radon and radon daughters. The third objective also concerns health, but in a much wider sense. The use of radon and radon daughters as tracers in the atmosphere is one of many tools helping to give us a better understanding of meteorological phenomena, such as air transport of pollutants.

Special emphasis in this investigation is given to:

- the time-dependent diffusion theory and its implications for radon exhalation measurements in closed volumes,
- the wire screen method to separate radon decay products unattached and attached to aerosols,
- continuous radon-in-air assessment with through-flow detectors,
- the problems connected with the measurement of very low concentrations of radon and radon daughters in maritime air.

1.4. NOMENCLATURE

In the text the specialized terms are usually defined as they occur. A few terms, however, need additional clarification, as they are often given different interpretations in the radon literature. Some of the abbreviations used are also explained below.

AMD is the Activity Median Diameter of an aerosol size distribution.

D is the diffusion coefficient, the proportionality constant in Fick's law of diffusion. For diffusive transport in porous material, effective diffusion coefficients can be defined (see Section 3.1 below).

EER, Equilibrium Equivalent Radon concentration, defined as the activity concentration of radon and daughters in equilibrium, having the same potential alpha energy concentration as the daughter mixture of interest.

emanation, by emanation in this report is meant the process by which radon atoms enter the pore space, thus becoming available for further transport in porous materials.

ϵ , the emanation factor, or emanation fraction, the fraction of the radon produced in a material that reaches the pore space.

E, exhalation, the phenomenon of radon atoms leaving a (macroscopic) material or sample. The exhalation can be expressed as the number of atoms or in activity units, and can refer to the mass, surface area etc. of the material in question.

exhalation rate, exhalation per unit time.

p, the porosity, is the ratio of pore-volume to bulk-volume of a material.

radon, ^{222}Rn , the radon isotope in the uranium decay series.

radon daughter, the decay products following ^{222}Rn in the natural uranium series.

RnD, short-lived radon daughters.

σ_{sp} , the light scattering coefficient of an aerosol. Measured in paper IV by instruments called nephelometers, sensitive to aerosol particles of diameters about 0.2-1 μm .

WL, Working Level, is an old unit of the quantity 'potential alpha energy concentration', equal to $1.3 \cdot 10^8$ eV per m^3 air, corresponding to an EER of 3750 Bq m^{-3} (≈ 0.1 pCi m^{-3}).

2. GENERAL PROPERTIES OF RADON AND ITS DAUGHTERS

2.1. RADON

The early suspicions that the 'emanation' was a gas belonging to the inert gases of the periodic table was soon verified. The physical properties of radon are given e.g. in the UNSCEAR report 1982 (UN82), and will not be repeated here. The chemical inertness of radon is an important feature both in atmospheric transport process (paper IV) and in connection with lung irradiation (paper III). The rare chemical compounds formed by radon, are reviewed by Stein (St83), but are of no consequence for this investigation. Like other inert gases, radon can be adsorbed by charcoal. The radon concentration apparatus used in paper IV exploits this principle. Charcoal adsorption is discussed in more detail in section 6 below.

2.2. RADON DAUGHTERS

The radon progenies, the radon daughters, are all isotopes of the metals polonium, bismuth or lead. As they are short-lived compared with the mother nuclide, they will soon reach radiation equilibrium in a closed system. (The prerequisite for radiation equilibrium between a radioactive daughter nuclide and its parent nuclide is either a long-lived parent or a constant supply of the parent.) In radiation equilibrium 1 becquerel of radon supports an equal activity of all four short-lived daughters. Each decay of ^{222}Rn in equilibrium with RnD generates 3 alpha particles in total.

In contrast to radon, the daughters are chemically and physically reactive. The behaviour of RnD:s in air is of special importance in papers III and IV and in these papers the phenomena of attachment to aerosol particles and plate-out onto surfaces have been described. The large difference in diffusivity between unattached RnD:s, and RnD:s attached to aerosol particles, is the basis of the wire screen separation method utilized in paper III, and the cause of the difference in deposition probability in the respiratory tract, which is relevant for the dose models used in paper III, and also discussed in section 5 below.

2.3. RADIATION DECAY AND EQUILIBRIUM

Radiation equilibrium in a closed system was briefly touched on in section 2.2. It should be pointed out that radiation equilibrium is not applicable to the airborne part of the activity in a closed system, such as in a zinc sulphide flask, due to plate-out. If, for instance, the mean plate-out rate constant for ^{218}Po is λ_p , and the only elimination of ^{218}Po is through radioactive decay and plate-out, the ratio between airborne ^{218}Po and ^{222}Rn is $\lambda_1/(\lambda_1 + \lambda_p)$, where λ_1 is the decay constant of ^{218}Po . The fraction $\lambda_p/(\lambda_1 + \lambda_p)$ of the ^{218}Po activity is deposited on surfaces by plate-out.

The half-life of the first long-lived daughter is 22.3 a (ICRP83). If we enclose the amount of 1 Bq of initially pure radon, several months later we will have more or less pure ^{210}Pb , but the decay rate of this radiolead will be very low, approximately $3.82/(22.3 \times 365) = 0.00047$ Bq. The low activity of long-lived radon progenies supported by a certain amount of radon or short-lived RnDs is of great practical consequence. The devices used to collect radon or short-lived daughters can normally be reused within a few hours after removal of the radon, as by then the deposited short-lived daughters have decayed. With increasing exposure (expressed, for instance, as time-integrated activity concentration, Bq s m⁻³) the background count rate will slowly increase, however, due to the accumulation of deposited long-lived daughters.

The exponential equations governing the decay and growth of the members of the decay series, are often in textbooks on nuclear physics referred to as 'Bateman equations' (Ba10, Ev55). A less well-known form of the Bateman equations, is the recurrence formula of serial decays, in which the number of atoms of the n:th daughter nuclide at time t, can be expressed as:

$$\text{Eq. 1} \quad N_n(t) = N_0(0) \cdot E_{0,1,2,3,\dots,n} \prod_{i=0}^{n-1} (\lambda_i)$$

if we, at time t=0, have a pure mother nuclide, indexed zero, and no branching. The functions E are built up from the simple function $E_0 = \exp(-\lambda_0 t)$.

$$E_{0,1} = \frac{E_0 - E_1}{\lambda_1 - \lambda_0} \quad E_{0,1,2} = \frac{E_{0,1} - E_{1,2}}{\lambda_2 - \lambda_0}$$

$$E_{0,1,2,\dots,n} = \frac{E_{0,1,2,3,\dots,n-1} - E_{1,2,3,\dots,n}}{\lambda_n - \lambda_0}$$

The full treatment of the recurrence formula is given by Hamawi (Ha71) and Scherpetz and Desrosiers (Sc81).

2.4. POTENTIAL ALPHA ENERGY

The alpha decay energy is of special relevance to radiation dosimetry (cf section 5 below). Concerning the radon daughter irradiation of the lung, it is the potential alpha energy of a daughter that is relevant, if we assume that the mean biological residence time of the daughters in the lung is long compared with the physical decay time. The assumption is not valid for the long-lived daughters and therefore the potential alpha energy of an atom is defined as the total alpha energy emitted during the decay of this atom along the decay series to ^{210}Pb (or ^{208}Pb for the thorium series) (ICRP 81). The potential alpha energies of the short-lived radon daughters are listed in Table 1.

TABLE 1.

Daughter No. i	Radionuclide	Potential alpha energy, p_i		Fraction of total, b_i
		nJ per Bq	MeV per Bq	
1	^{218}Po (RaA)	0.579	3,61 ₄	0.104
2	^{214}Pb (RaB)	2.85 ₇	17,8 ₃₃	0.514
3	^{214}Bi (RaC)	2.12 ₁	13,2 ₄₁	0.382
4	^{214}Po (RaC')	2.9 10^{-7}	1.8 10^{-3}	< 10^{-7}
	Total	5.55 ₈	34,6 ₈₉	1.000

Having an arbitrary mixture of radon daughters in air, the potential alpha energy concentration, C_{pot} , is obtained as the sum

$$\text{Eq. 2} \quad C_{\text{pot}} = \sum_{i=1}^4 p_i C_i \quad [\text{J m}^{-3}]$$

where the C_i 's are the activity concentrations of the individual daughters. The contribution from ^{214}Po in Eq. 2 is so small, that for all practical purposes, the upper index of the sum can be set to $i=3$. The equilibrium equivalent radon concentration, EER, of a non-equilibrium mixture, C_i , is simply the radiation equilibrium mixture of short-lived daughters having the same potential energy concentration, C_{pot} , that is

$$\text{Eq. 3} \quad C_{\text{pot}} = \sum_{i=1}^3 p_i C_i = \sum_{i=1}^3 p_i \cdot \text{EER}$$

From equation 3 we easily obtain (qf Table 1):

$$\text{Eq. 4} \quad \text{EER} = \sum b_i C_i = 0.104 C_1 + 0.514 C_2 + 0.382 C_3 \quad [\text{Bq m}^{-3}]$$

The radon daughter equilibrium factor F, with respect to potential alpha energy, is defined as

$$\text{Eq. 5} \quad F = \frac{\text{EER}}{C_{\text{Rn}}}$$

where C_{Rn} is the radon concentration in air.

3. RADON AND RADON DAUGHTER TRANSPORT

The intention of this section is to briefly introduce the fundamental principles of radon sources and radon and radon daughter transport. Relevant aspects of both microscopic radon migration (paper I) and of large-scale transport (paper IV) are presented.

3.1. RADON MIGRATION IN POROUS MEDIA

In paper I the radon transport in a material with air-filled pores is theoretically described as a pure diffusion process. To advance the understanding of the theoretical part of paper I, the transport processes and associated parameters will be discussed in more detail in this section.

The literature in the field is extensive. The interested reader is referred to the comprehensive reviews by Tanner (Ta64, Ta80), covering geological literature on radon migration in soil, listing more than 450 references, and to the review study of radon transport in building materials performed by the National Bureau of Standards (Co81), giving 174 references.

In the porous medium the pores are filled with a 'fluid', e.g. air, water etc. The migration of radon atoms in the pores can be divided into two broad categories, either a movement with the fluid or a movement with respect to the fluid. An example of the former is a pressure gradient driving the fluid, bringing radon with it, and an example of migration relative to the fluid is diffusive transport. The porosity, p , of the material is an important parameter, defined in the glossary. In paper I, the definition of p assumes that the fluid is air throughout.

Even in the earliest radon measurements in soil it was noticed that only a fraction of the radon produced from ^{226}Ra reached the pore space. Meyer and Schweidler (Me27) give the range $\epsilon = 5 - 17\%$ referring to Satterly's measurements of soil gas 1911-12. In most materials, the grain size is much larger than the range of the recoiling radon atom, giving low values of ϵ . Water or other liquids in the pores increase the emanation fraction ϵ , as experimentally observed by several authors (see Co81 for references) including ourselves (Sa82). The increased ϵ -value is explained by the higher probability of a radon atom stopping in a water-filled pore than in an air-filled pore. From this it is also understood that the 'water effect', promoting high values of ϵ , is depen-

dent on pore size. The recoiling ^{222}Rn atom has an energy of 0.10 MeV and a range of about 63 μm and 0.1 μm in air and water respectively (Ta80). Emanation factors in the range $\epsilon = 1 - 70\%$ have been reported (Wi80, Ta80). It should be added that in certain minerals the ^{226}Ra atoms may be inhomogeneously situated in the grains. If ^{226}Ra atoms are superficially concentrated, high emanation factors may be obtained, even without moisture-filled pores.

The exhalation of radon from a macroscopic body is a two-step process, first the liberation of the radon atoms to the pore volume, secondly the migration of the radon atoms to the surface of the body. In our opinion, it is therefore pertinent to define a special term, the emanation factor ϵ , as above, applicable only to the first step in the exhalation process. The well established and well defined quantity, 'emanation power' or 'escape-to-production ratio' is equal to the quotient radon activity liberated from the material (exhaling), by the radon activity formed in the material, during a time unit (Ta80). Using this definition our 'emanation factor' is the same as the emanation power of the grains or at least very small bodies, small enough not to appreciably delay the liberation of radon from the body. The term 'emanation power' is awkward, involving both steps of the exhalation, and is thereby dependent on the macroscopic dimensions of the material. The term is definitely unsuitable during the non-steady-state conditions discussed in paper I.

Consider a binary system of radon and air. If a radon concentration gradient exists, there will be a net transport of radon in the direction of decreasing concentration. This transport phenomenon is called diffusion. Nearly 130 years ago Fick put earlier qualitative observations of diffusion on a quantitative basis by introducing what is now known as Fick's law. In one dimension it says:

$$\varphi = -D \frac{dn}{dz} \quad [\text{m}^{-2}\text{s}^{-1}]$$

where φ is the (particle) fluence rate, D is the diffusion coefficient ($\text{m}^2 \text{s}^{-1}$), n is the concentration of atoms (m^{-3}), and z is the distance (m). Following the International Commission on Radiation Units and Measurements (ICRU80), we prefer the name particle fluence rate for φ , instead of the common but equivocal name 'flux density'. If we express the concentration as an activity concentration, C , we have

Eq. 6 $j = -D(dC/dz)$

where $j = \varphi \cdot \lambda_f$ is the activity fluence rate, and $C = \lambda_f \cdot n$. λ_f is the physical decay constant of radon. If we add a pressure gradient dp/dz , the activity fluence rate will change, due to the air movement. A term, $v C$, should be added to equation 6, where the air volume current v ($m^3 s^{-1} m^{-2}$) can be found from Darcy's law, $v = -(k/\eta) dp/dz$ (Wi80, Co81), where k is the permeability coefficient, and η the dynamic viscosity. In paper I, both pressure- and thermal (causing thermal diffusion) gradients are assumed to be non-existent.

If the air is homogeneously distributed in space, the diffusion coefficient in Eq. 6 is the molecular diffusion coefficient, D_m . In a porous medium the diffusive air flow is restricted and we have to use an effective diffusion coefficient. Failure of authors to distinguish between different diffusion coefficients has led to confusion in the literature, as pointed out by Culot (Cu76). The aim of the following is to deduce the differential diffusion equation and to clarify the relationships between the molecular diffusion coefficient, D_m , and the different effective diffusion coefficients which can be defined.

Let us consider a homogeneous and stagnant medium, e.g. air, where we have an activity concentration of radon, C , and a concentration gradient ∇C . To be general we write Fick's law in three dimensions

$$-D_m \nabla C(x,y,z) = \vec{j}_m$$

where

D_m is the true molecular diffusion coefficient

∇ is the nabla operator

$\vec{j}_m$ is the vectorial activity fluence rate.

If we now introduce infinitely thin but non-permeable microscopic walls in the medium, the diffusive flow will be obstructed due to the tortuosity of the 'maze'. For the same concentration gradient the activity fluence rate decreases by a factor h , the 'tortuos' factor

$$\text{Eq. 7} \quad -h D_m \nabla C(x,y,z) = h \vec{j}_m$$

Penman has published values of h in the range 0.5 - 0.75 (Pe40). The second step is now to make the walls thicker, without changing the hypothetical maze in any other respect. Only a fraction, the porosity p , of the total volume, is air, but equation 7 is still valid, if the fluence rate refers to an unoccluded area and C refers to the activity concentration in the pore volume. If f denotes the

unoccluded fraction of a cross sectional area, the activity fluence rate, $\bar{j}$, referring to a full area is equal to $f h \bar{j}_m$, and we have

$$- f h D_m \nabla C(x, y, z) = - D_e \nabla C(x, y, z) = \bar{j}$$

which is Fick's law for a porous medium. D_e is called the effective diffusion coefficient, designated by k_e in Culot's paper (Cu76).

The differential equation for diffusion in a porous material (Eq. 1 in paper I) is based on the validity of Fick's law together with a balance calculation for the rate of change in radon activity, per bulk volume, as shown in the following deduction.

i/ The increase, g , due to decaying radium atoms can be written $g = C_{Ra} \rho \lambda_f \epsilon$, where C_{Ra} is the specific radium concentration ($Bq\ kg^{-1}$) of the sample, ρ the bulk sample mass density, λ_f the decay constant of radon and the emanation factor.

ii/ The decrease due to decaying radon atoms is simply $\lambda_f p C$ per unit time, as the number of decaying radon atoms per bulk unit volume is $p C$. C is the radon concentration in the pore volume, and p the porosity.

iii/ The change due to radon diffusion is generally $\iint_A \bar{j} \cdot \bar{da}$, where da is a surface element on the closed volume V . (The surface enclosing V is denoted by A). The change may be rewritten as a volume integral, using Fick's law and the theorem by Green (see any textbook on vector analysis)

$$- \iint_V \bar{j} \cdot \bar{da} = \iint_V D_e \nabla C(x, y, z) \cdot \bar{da} = D_e \iiint_V \nabla^2 C(x, y, z) dv$$

The change of radon activity in the closed volume V per unit time can now be written

$$\iiint_V p \cdot \frac{\partial C}{\partial t} dv = \iiint_V (g - \lambda_f p C) dv + D_e \iiint_V \nabla^2 C(x, y, z) dv$$

In the limit of very small volumes, the variability in space can be neglected and therefore

$$\text{Eq. 8a} \quad \frac{\partial C}{\partial t} = g/p - \lambda_f \cdot C + \frac{D_e}{p} \cdot \nabla^2 C$$

Introducing the maximum of C in an infinite sample, $C_m = C_{Ra} \cdot \rho_e / p$, and the diffusion length $L = (D_e / \lambda_f p)^{0.5}$, the diffusion equation can be expressed as

Eq. 8b

The deduction of the diffusion equation and Fick's law applicable to a porous medium, laid out above, does not require any special relation between the porosity p and the unoccluded fraction f . The effective diffusion coefficient used in paper I is defined as $D = D_e / p = f h D_m / p$. Another and more common way to define an effective diffusion coefficient is as $D_{eff} = h D_m$. This latter definition leads to the diffusion equation

$$\frac{\partial C}{\partial t} = \frac{g}{p} - \lambda_f \cdot C + \frac{f}{p} D_{eff} \nabla^2 C$$

and Fick's law takes the form

$$-f D_{eff} \nabla C = \bar{j}$$

To avoid the complication of working with both porosity, p , and unoccluded fractional area, f , as independent parameters, it is customary to assume equality between them. When presenting theories or experimental results in the field of diffusion in porous materials, it is important to clarify which one of the effective diffusion coefficients one is referring to. This can be done unambiguously by giving the theoretical relation between the effective coefficient and the molecular diffusion coefficient, as done above and in paper I.

The objective of paper I is to apply the diffusion equations to exhalation measurements on enclosed samples, and for that purpose we need both the time-dependent and the steady-state ($\partial C / \partial t = 0$) solution of Eq. 8. The solutions will have different forms depending on initial and boundary conditions. An infinite slab of thickness $2d$ in free space, has the same areal exhalation rate as a sample of thickness d in an open canister. The proof of this is given in paper I. It is therefore sufficient in the case at hand to work with the one-dimensional solutions of Eq. 8.

The time-dependent pore concentration in a sample closed at time $t = 0$, initially exhaling into free space, is given in Eq. 7 in paper I. The corresponding exhalation is (a radon-tight vessel is assumed, that is $\gamma = 1$)

$$\text{Eq. 9} \quad E_{\ell}(t) = -D_e \left. \frac{\partial C(z,t)}{\partial z} \right|_{z=d} = E_{\ell}(\infty) + C_m \frac{2 \tanh(d/L) \cdot \exp\{-\lambda_f t\}}{d/L} + \sum_{i=1}^{\infty} \frac{\lambda_f \ell y_i^2 L^2 \exp\{-(y_i L/d)^2 \lambda_f t\}}{(1 + \alpha + \alpha^2 \cdot y_i^2)(d^2 + y_i^2 L^2)}$$

where y_i are the non-zero positive roots of $-\alpha y_i = \tan y_i$, and $E_{\ell}(\infty)$ the bound steady state exhalation (Eq. 5 in paper I). The first ten y_i values are given in Table 2 for three different outer- to inner volume ratios, α .

Table 2.

	$\alpha=0.1$	$\alpha=1$	$\alpha=10$		$\alpha=0.1$	$\alpha=1$	$\alpha=10$
y_1	2.8631	2.0287	1.6320	y_6	17.791	17.336	17.285
y_2	5.7611	4.9131	4.7335	y_7	20.868	20.469	20.425
y_3	8.7089	7.9786	7.8667	y_8	23.958	23.604	23.566
y_4	11.703	11.086	11.005	y_9	27.058	26.741	26.707
y_5	14.734	14.207	14.144	y_{10}	30.166	29.879	29.849

The solution of the time dependent concentration in Eq. 7 (I) has been presented by Krisiuk (Kr71) and Lamm (La84). (In the English translation of Krisiuk's paper available to us, the minus sign in the transcendental equation is missing). The practical implications of the theoretical equations given above will be discussed in section 7 below. Only a few comments to facilitate the interpretation will be given here.

The relation between free- and steady-state bound exhalation for a vessel with a leakage factor γ is (Eq. 6 in paper I)

$$\text{Eq. 10} \quad \frac{E}{E_{\ell}(\infty)} = \frac{\coth(d/L) + \frac{pL}{\gamma \ell}}{\coth(d/L)} = 1 + \frac{pL \cdot \tanh(d/L)}{\gamma \ell}$$

$$= \begin{cases} 1 + pL/\gamma \ell & \text{if } d \gg L \\ 1 + 1/\gamma \ell & \text{if } d < 0.5 L \end{cases}$$

The maximum pore concentration in a free sample is solely determined by the thickness to diffusion length ratio, d/L . Introducing an enclosure will increase this maximum. An increased ventilation expressed as a relative leakage, has the same effect on the steady-state bound exhalation, as increasing the height (l) of the outer volume. Increasing the outer volume or the ventilation rate will raise the steady-state exhalation rate. The physical explanation is easy; to support the larger radon 'elimination' in the outer volume, we need a larger exhalation.

In Eq. 7 (I) the two first terms on the right-hand side are time-independent. They correspond to the steady-state exhalation, and involve a depth, (z), dependence. The third term, on the other hand, is only time-dependent, and the first three terms seen together describe how the radon concentration within the sample increases with time in a congruent fashion, without changing the concentration-depth gradient (i.e. exhalation). All parameters describing the change in the concentration gradient with time, are gathered in the last term, $G(z,t)$, of Eq. 7 in paper I. For thin samples ($d < L$) there is a fairly rapid change from free exhalation, at time zero, towards bound exhalation. The time it takes upon closing for the exhaling system to reach 95 % of its final steady-state exhalation value is given in Table 1 of paper I.

3.2. DISPERSION OF RADON AND RADON DAUGHTERS IN THE ATMOSPHERE

As discussed in paper IV, the continents act like large areal sources of radon, exhaling at a rate approximately two orders of magnitude greater than the oceans. The radium content of the atmosphere is exceedingly small, and the only radon source of importance in the atmosphere budget is the ground.

The exhaling radon will be dispersed in the mixing layers of the atmosphere, showing a decreasing concentration with altitude. In this mixing layer (called the atmospheric boundary layer in meteorology), typically of 1 km height, the radon will be eliminated mainly by physical decay. The physical decay is also the dominating removal process for the short-lived daughters, while precipitation and dry deposition removal are the most important elimination routes for ^{210}Pb ($T_{1/2}=22$ a). The aim of this section is to give a brief overview of how radon and radon daughters participate in atmospheric processes, and the intention is that such information will be relevant to paper IV.

3.2.1 THE BEHAVIOUR OF OUTDOOR RADON

The ground can be treated as a porous source medium, just as in section 3.1. The detailed treatment in 3.1, describing the diffusive exhalation is, however, in this case less meaningful. Local meteorological conditions cause large variations in the radon exhalation rate. The transport of radon to the ground surface is caused by a mixture of concentration-, temperature- and pressure gradients as well as water flow. The transport modes through both small-sized openings in granular soil and larger fissures vary greatly with time and space, making anything but large-scale and long-term models difficult to apply. Once the radon concentration gradient dC/dz , the pressure gradient dp/dz , the temperature gradient dT/dz , and the water flow in the ground are known, the radon transport and exhalation can be theoretically calculated. Equations analogous to equation 4 are used, (Fick used, for example, Fourier's law of heat energy fluence rate, which is proportional to the temperature gradient, as a pattern, in formulating his law of diffusion), but such a penetrating analysis is beyond the scope of this investigation, and is of little relevance to paper IV.

In terms of the large-scale considerations relevant during the Ymer expedition, the ground can be seen as a radon generator, supporting an air concentration of radon. This air concentration, over land, is the source of interest in paper IV. In a small air volume above ground (height z), a radon activity balance equation similar to Eq. 8 above can be formulated. Some modifications have to be made, however. As mentioned above, the radium concentration of the air is insignificant, thus the source term, g , in Eq. 8 is zero. The homogeneous diffusion coefficient, D_m , should be used in Fick's law (cf page 16) and we have for the radon activity concentration, C , in air

$$\text{Eq. 11} \quad \frac{\partial C}{\partial t} = -\lambda_f C + D_m \nabla^2 C$$

But we must further modify Eq. 11, as molecular diffusion is not a very active transport process compared to the turbulent action of the air. The turbulent transport of radon into upper layers can mathematically be treated as a diffusion process, by introducing an eddy (turbulence) diffusion coefficient D_t in Eq. 11. The equivalence of molecular diffusion and turbulence diffusion is not complete, however, and D_t cannot, in more realistic situations, be treated as a constant. To take this into account the stationary balance equation may be written in one dimension, ($D_t \gg D_m$) as (Is62)

$$\text{Eq. 12} \quad D_t \cdot \frac{d^2 C}{dz^2} + \frac{dD_t}{dz} \cdot \frac{dC}{dz} - \lambda_f C = 0$$

The one-dimensional form requires that the radon concentration is constant in the horizontal plane, which is possible only in the middle of larger regions of constant areal exhalation rate. In paper IV we have used Eq. 12 to assess the radon concentration over the ocean surface. The two dominant factors, determining the radon content in surface air are the exhalation rate and the upward transport by turbulent agitation. As exhalation rate and turbulent mixing exhibit both seasonal and diurnal variations, so does the radon concentration. References to this aspect of radon in outdoor air have been given in paper IV. A typical diurnal variation is a factor of about 2 (ratio max. to min.) but it should be observed that in individual cases precipitation is a complicating factor, causing large variations in the exhalating rate. The daily variations of airborne daughter activities at ground level are even greater.

In addition there are also uncertainties concerning the transport of radon from the source (the continents) to the polar region. Several long-range transport models exist (cf e.g. the review paper by Fisher, 1983), applicable to primary 'pollutants' like SO_2 and radon, but these models make reliable predictions only over long time periods, about a year or more. The models agree to within a factor of two in this long-time perspective, but to cite Fischer 'no such agreement is possible, nor is likely to be achieved in the near future, between models and daily measurements'. The citation supports the decision of the authors of paper IV, to keep the discussion of the Ymer day-to-day results mainly on a qualitative level.

3.2.2. THE SHORT-LIVED RADON DECAY PRODUCTS IN OUTDOOR AIR

Airborne radon is the only significant source of short-lived daughters in the mixing layer. The fates of daughters are more diverse compared with radon, as they are influenced by several different scavenging effects. Of these, as discussed in paper IV, only elimination by precipitation may be fast enough to be comparable to the half-lives of the daughters. The consequence of this is that, except for precipitation periods and few hours thereafter, the short-lived daughters are in approximate equilibrium with radon. This does not, however, necessarily apply in a short time perspective at heights of up to a few metres above ground. At these small 'altitudes' temporal variations in the radon exhalation combined with fast changes in turbulence mixing, interact in a complicated manner. Short-time deviations of duration, a few hours, from the close-to-equilibrium conditions, should be expected during changing atmospheric conditions. For further references the interested reader is referred to Schuman (1972) and Gogolak & Beck (1980). The latter authors also stress the difficul-

ties in obtaining realistic solutions to the radon daughter balance equations at height z

$$\text{Eq. 12} \quad D_t \cdot \frac{d^2 C}{dz^2} + \frac{dD_t}{dz} \cdot \frac{dC}{dz} - \lambda_f C = 0 \quad (\text{Radon})$$

$$\text{Eq. 13} \quad D_t \cdot \frac{d^2 C_i}{dz^2} + \frac{dD_t}{dz} \cdot \frac{dC_i}{dz} + \lambda_{f,i-1} \cdot C_{i-1} - (\lambda_{f,i} + \lambda_r) C_i = 0 \quad (\text{RnD})$$

where the index i denotes the i :th daughter nuclide and λ_f and λ_r are the physical decay and removal rate constants respectively. Below about 100 metres the turbulence diffusion coefficient D_t decreases proportionally to z^a , where a is between 1.2 and 1.5 (Go80). Depending on the atmospheric stability D_t takes different absolute values (cf Ja63).

In equation 13 short-lived daughters attached and unattached to the atmospheric aerosol are combined together. It is not the objective of this investigation to elaborate on the radon daughter attachment process in outdoor air. If interested, the reader should consult the monograph of Junge (Ju63) or relevant references given in paper IV. But it should be stressed here, as in paper IV, that the radon daughter activity is predominantly in the attached 'phase'. The disequilibrium between RnDs and radon in the atmosphere during dry and precipitation periods have been discussed by Gat et al (Ga66). The fact that most RnD activity is attached to aerosol particles facilitates the sampling, as unattached RnDs would easily be lost in any pipes or tubing preceding the sampling filter.

3.2.3 THE LONG-LIVED RADON DAUGHTERS IN OUTDOOR AIR

The intention in this section is to summarize the main features of the long-lived products' behaviour in outdoor air. The half-lives of ^{210}Pb , ^{210}Bi and ^{210}Po are 22 a, 5.8 d and 138 d respectively, which are much longer, or comparable to the time-scale for scavenging by dry deposition, sedimentation, rain-out etc. Some insight into the elimination processes is gained starting from the unrealistic steady-state assumption of a constant radon concentration and an identical removal rate constant, λ_r , of all radon daughters. If we modify Eq. 1 to include a 'branching' factor f , and replace the physical decay constant with an 'effective' decay constant, $\lambda_i' = \lambda_i + \lambda_r$, we obtain

$$\text{Eq. 14} \quad N_n(t) = N_0(0) \cdot E'_{0,1,2,\dots,n} \cdot \prod_{i=0}^{n-1} (f_{i+1} \lambda_i')$$

where f_{i+1} equals the ratio $\lambda_i / (\lambda_i + \lambda_r)$, that is the fraction of the i :th daughter atoms decaying to daughter $i+1$. The removal rate constant, λ_r , is assumed to be zero for radon. The E' function is defined as $E'_i = \exp(-\lambda'_i t)$, in analogy with the derivation given earlier. Having a constant production $\dot{N}_0$ (s^{-1}) of radon atoms starting at time $t = 0$, we can find the number of atoms of the n :th daughter at time t , by an integration of Eq. 14

$$\begin{aligned} \text{Eq. 15} \quad N_n(t) &= \int_0^t \dot{N}_0 E'_{0,1,2,\dots,n} \prod_{i=0}^{n-1} (f_{i+1} \lambda'_i) dt = \\ &= \dot{N}_0 \prod_{i=0}^{n-1} (f_{i+1} \lambda'_i) \cdot I'_{0,1,2,\dots,n} \end{aligned}$$

The I' function is calculated recurrently:

$$\begin{aligned} I'_0 &= \int_0^t E'_0 dt = \frac{1 - e^{-\lambda'_0 t}}{\lambda'_0} \quad I'_{0,1} = \frac{I'_0 - I'_1}{\lambda'_1 - \lambda'_0} \\ I'_{0,1,2,\dots,n} &= \frac{I'_{0,1,2,\dots,n-1} - I'_{1,2,3,\dots,n}}{\lambda'_n - \lambda'_0} \end{aligned}$$

In the limit $t \rightarrow \infty$ the integrated function I' takes the form

$$\text{Eq. 16} \quad \lim_{t \rightarrow \infty} I'_{0,1,2,\dots,n} = \frac{1}{\prod_{i=0}^n (\lambda'_i)}$$

and in the steady state ($t \rightarrow \infty$) we have, from Eq. 15 and Eq. 16

$$N_n(\infty) = \dot{N}_0 \prod_{i=0}^{n-1} (f_{i+1} \lambda'_i) \cdot \frac{1}{\prod_{i=0}^n (\lambda'_i)} = \frac{\dot{N}_0}{\lambda_n + \lambda_r} \prod_{i=0}^{n-1} (f_{i+1})$$

The ratio of the n :th daughter activity to the radon activity at steady state, can then be written

$$\begin{aligned} \text{Eq. 17} \quad \frac{C_n(\infty)}{C_0(\infty)} &= \frac{\lambda_n \cdot N_n(\infty)}{\lambda_0 \cdot N_0(\infty)} = \frac{\lambda_n \cdot N_n(\infty)}{\dot{N}_0} = \frac{\lambda_n}{\lambda_n + \lambda_r} \prod_{i=0}^{n-1} (f_{i+1}) = \\ &= \frac{\lambda_1}{\lambda_1 + \lambda_r} \cdot \frac{\lambda_2}{\lambda_2 + \lambda_r} \cdot \dots \cdot \frac{\lambda_n}{\lambda_n + \lambda_r} \end{aligned}$$

If we compare the n:th daughter activity concentration with its mother, the preceding nuclide n-1, we have

$$\text{Eq. 18} \quad \frac{C_n(\infty)}{C_{n-1}(\infty)} = \frac{\lambda_n}{\lambda_n + \lambda_r}$$

The mean residence time for aerosols, $T_r = \lambda_r^{-1}$, in the outdoor environment is of the order of a few days, or more. For the short-lived radon daughters ($i = 1 \rightarrow 4$) therefore, $\lambda_r \ll \lambda_i$ is a good approximation.

Knowing the activity ratios from measurements, it is possible to calculate the mean residence time, T_r , from Eq. 17 or Eq. 18. To obtain accuracy in such a calculation the removal rate constant should be of the same order as, or larger than, the physical decay constant. This criterion for λ_r is fulfilled for the long-lived daughters. It is difficult, however, to comply with the demands on the equilibrium condition, on which Eq. 17 is based. As mentioned earlier the radon exhalation from the ground exhibits diurnal variations, the removal coefficient is subjected to large changes during precipitation, and so on. In addition the calculation of aerosol residence time will be unreliable due to mixing of air masses having different scavenging histories, and due to the existence of other radon daughter sources than airborne radon. Moore et al (Mo75) for instance estimate that over 50% of the ^{210}Po activity in the surface air over the west-central USA originate directly from resuspended soil materials.

It is of particular interest in our case to see what is happening to the long-lived daughter activity content of an air mass moving out over the sea. To idealize the initial conditions, let us assume that the radon daughter system is in a steady state with a constant removal rate constant of λ_r , before leaving land which also prevails over the sea. Further, the assumption is that radon is removed from the air only by physical decay and that the dispersion of radon and its daughters in the sea air is identical except for the specific aerosol activity removal, described by λ_r . At time $t = 0$ this idealized air mass leaves the continent for the sea. Following the fate of this air mass, remaining over the sea, we observe that the exhalation radon source is removed (the exhalation of radon from the sea is negligible), and that initially the ratio between, for example, ^{210}Pb and radon is approximately equal to $\lambda_4 / (\lambda_4 + \lambda_r) \simeq \lambda_4 / \lambda_r$, where λ_4 is $8.5 \cdot 10^{-5} \text{ d}^{-1}$. What happens to the activity ratio $^{210}\text{Pb}/^{222}\text{Rn}$ for $t > 0$? To answer this question qualitatively we can use our knowledge about equilibrium in radioactive decay series. If the mean residence time in air, $\lambda_r^{-1} = T_r$, for ^{210}Pb is longer than that for radon, 5.5 days, the ratio $^{210}\text{Pb}/^{222}\text{Rn}$ will increase and become infinite, if T_r is slightly less than 5.5 days, there is still an increase but the ratio $^{210}\text{Pb}/^{222}\text{Rn}$ will take on a finite value. When T_r

is much less than 5.5 days we have the conditions of the long-lived mother and short-lived daughter, which means that the daughter will be in radiation equilibrium with its mother nuclide. In our case only the airborne daughters are of interest, not the ones removed, so the ratio of airborne ^{210}Pb to ^{222}Rn will be equal to approximately λ_4 / λ_r , if $\lambda_r^{-1} \ll 5.5$ days. To see quantitatively the time development of the activity in the air mass dealt with above we can use equation 14. We have to modify it though, to take into account that we initially, at time $t = 0$, have daughter activities present. This is done by a summation over all nuclides preceding and including the daughter of interest, and applying the relevant E' function to each of the daughters.

$$\text{Eq. 19} \quad N_n(t) = \sum_{i=0}^n N_i(0) \prod_{j=1}^{j=n-1} (f_{j+1} \lambda'_i) \cdot E'_{i,i+1,\dots,n}$$

To facilitate the interpretation, we can write the compact formula of Eq. 19 explicitly, remembering that an empty factorial product ($i=n$) is equal to 1.

$$N_0(t) = N_0(0) E'_0$$

$$N_1(t) = N_0(0) f_1 \lambda'_0 E'_{0,1} + N_1(0) E'_1$$

$$N_2(t) = N_0(0) f_1 f_2 \lambda'_0 \lambda'_1 E'_{0,1,2} + N_1(0) f_2 \lambda'_1 E'_{1,2} + N_2(0) E'_2 \quad \text{etc.}$$

The theoretical results of calculations following Eq. 19 are shown in Figure 11 in paper IV, for different aerosol residence times. As the aerosol residence time used is much longer than the half-lives of the short-lived daughters, the influence of these has been neglected.

It is obvious, in spite of the idealized conditions on which Figure 11 (IV) is based, that it is fundamentally wrong to use quotients of activity (e.g. $^{210}\text{Pb}/^{222}\text{Rn}$ or $^{210}\text{Bi}/^{210}\text{Pb}$) to calculate mean aerosol residence times T_r , unless T_r is about 1 day or shorter, or unless both initial activity values and the time since the air mass left the continent, are known. Even the ratio $^{210}\text{Bi}/^{210}\text{Pb}$, proposed by Martell and Moore (Ma74) as the best ratio to use for estimates of tropospheric residence times, cannot be used with confidence in maritime air. If the air mass system is interpreted as a steady-state system, there is a high probability of an overestimation of the mean residence time, if

the true residence time is longer than, say, two days. In paper IV the aerosol residence time is estimated to between 4 and 7 days, using the grand mean $^{210}\text{Pb}/^{222}\text{Rn}$ ratio for the Ymer-80 expedition and the calculations resulting from Equation 19 (cf Figure 11 IV). (The steady state equilibrium formula yields an apparent aerosol residence time of 12 days).

4. RADON AND RADON DAUGHTERS IN INDOOR AIR

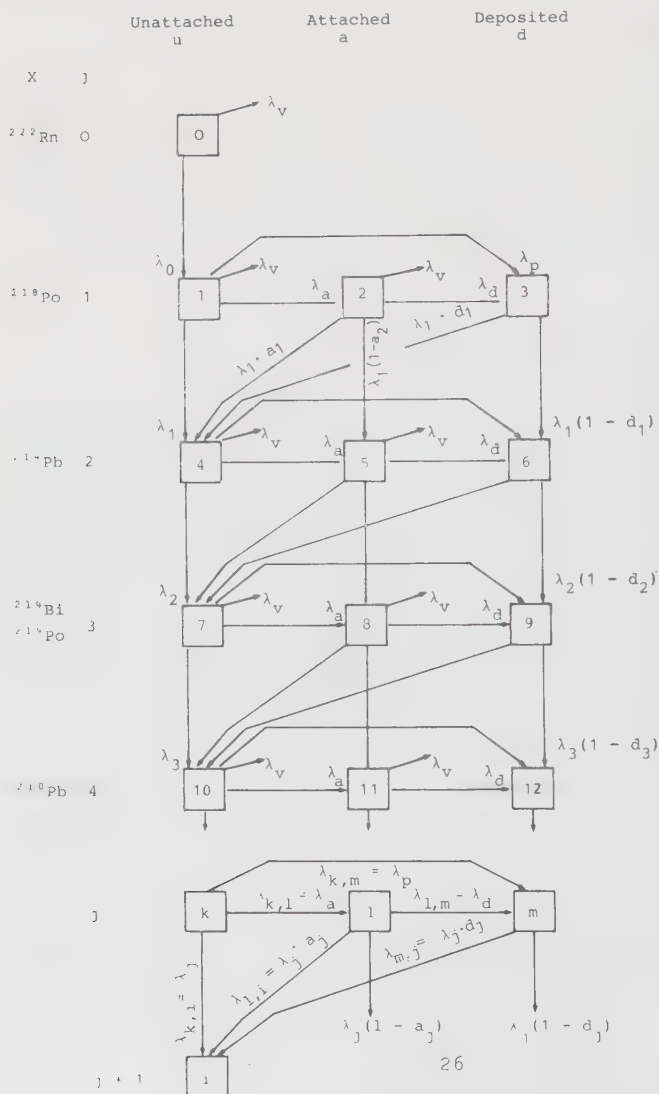
In connection with the air-cleaning paper (paper III) it is relevant to give a short overview of the radon- and radon daughter levels and behaviour in indoor environments. For a more comprehensive review the reader is recommended to consult the latest UNSCEAR report (UN82) and references therein. For radon levels in the Nordic countries a joint report from the radiation protection institutes in these countries has recently become available (NRP83). A special issue of Health Physics (HP83), published in August 1983 is solely devoted to indoor radon. Since the appearance of the UNSCEAR report, a review paper entitled 'Radon and Thoron in Buildings' has been published by Meggitt (Me83). The great interest in recent years in indoor radon has been caused by the increased awareness that radon irradiation, through the inhalation of short-lived daughters, is a substantial, and in some countries a dominant, part of the 'natural' radiation environment. Some groups of people in the Nordic countries actually receive 'doses' higher than the dose limit for radiation workers (NRP83). (In section 5 below, radon dosimetry will be discussed).

4.1. THE RADON- AND RADON DAUGHTER BUDGET IN A ROOM.

In general the potential sources of radon are materials of high ^{226}Ra content or fluids originating from such materials. The source may be the soil beneath a house, construction material, or water or gas supplied to a house. In part 2 above the radon migration in porous materials was discussed. The transport processes mentioned there, porous diffusion, pressure- or temperature-induced flow of air and water are all relevant to radon in indoor areas, but in individual cases one route of entry may be dominant. Large variations with time in the radon entry rate (Bq s^{-1}) are common if the radon source is the soil or the water supply. In the soil case, an important factor is the pressure difference between soil and house, excess pressure in the house may prohibit radon entry more or less completely. The fresh water consumption in a house may change rapidly, and with it the radon, entering the house dissolved in the water. We shall not elaborate on the entry routes any further, but conclude by saying that the very extreme indoor radon concentrations in Sweden are found in houses where the radon emanates from the subsoil, and not the building materials, (UN82, NRP83).

Once inside a confined space such as a dwelling the radon will be removed by decay or ventilation. Theoretical calculations on the radon- and radon daughter

behaviour in an indoor environment, are facilitated by the fact that the influence of meteorological parameters, which are hard to quantify, is less than in the outdoor case. On the other hand, further complexity is added to the indoor case as a distinction between daughter activity attached and unattached to aerosol particles has to be made when studying the radon inhalation effects. As usual the external gamma irradiation from the daughters is considered unimportant compared with the alpha irradiation of the respiratory organs. The radon daughter behaviour and elimination paths in a room are best illustrated by a compartment model (Figure 1).



Only the airborne daughters are a direct irradiation risk. The activity deposited on a surface must be kept trace on as it may be airborne again via recoil effects. We can use the compartment model in Figure 1 to investigate the fate of the radon daughters in a room. It is assumed that the net infiltrating radon is the only source of daughter activity. The j :th radionuclide can be in different compartments (numbered from zero to twelve) as unattached, attached or deposited activity. The airborne activity is ventilated away in a first order process with the rate constant λ_v . When the j :th daughter attached to the aerosol disintegrates, there is a probability, a_j , that the daughter will become unattached. The corresponding probability for deposited activity decaying, is denoted by d_j . The four rate constants for physical decay, attachment, deposition of attached, and deposition of unattached activities are called λ_j , λ_a , λ_d and λ_p respectively.

Even without air filtration, it is obvious that the radon daughter 'budget' in a room is rather complex. If we wish to take filtration removal into account, we may exchange the ventilation rate constant, λ_v , with an effective removal rate constant, $\lambda_v + \lambda_c$, for airborne radon daughters. The effective filtration rate constant, λ_c , may be different for attached and unattached daughters, due to different collection efficiencies of the filter media used. Introducing air cleaning will also affect the deposition rate constants, increasing the probability of plate-out, due to the increased air turbulence.

The degree of non-equilibrium at steady state between the airborne radon daughters and radon is of special interest. Describing the degree of non-equilibrium for the j :th daughter by its activity ratio to radon, g_j , the following equation for attached activity can be derived

$$\text{Eq. 20} \quad g_{j,a} = \frac{A_{j,a}(\infty)}{A_0(\infty)} = \frac{1}{\lambda_j + \lambda_d + \lambda_v} \left\{ \lambda_a g_{j,u} + \lambda_j (1 - a_{j-1}) g_{j-1,a} \right\}$$

where the indices a and u stand for attached and unattached daughters respectively. The corresponding expression for the unattached activity ratio is

$$\text{Eq. 21} \quad g_{j,u} = \frac{A_{j,u}(\infty)}{A_0(\infty)} = \frac{\lambda_j}{\lambda_j + \lambda_a + \lambda_p + \lambda_v} \cdot \left\{ g_{j-1,u} \left(1 + d_{j-1} \frac{\lambda_p}{\lambda_{j-1}} \right) + g_{j-1,a} \left(a_{j-1} + d_{j-1} \frac{\lambda_d}{\lambda_{j-1}} \right) \right\}$$

As all radon activity is considered unattached, $g_{0,u} = 1$ and $g_{0,a} = 0$, giving the initial conditions for Eq.s 20 and 21.

Another quantity of interest is the fraction, $h_{j,u}$, of the airborne daughter j , which is unattached. This can be calculated as the ratio $g_{j,u}/(g_{j,u} + g_{j,a})$ from

Eq. 20 and Eq. 21. The unattached fraction for ^{218}Po , $h_{1,u}$, is given by a simple formula, as the detachment processes, have no effect

$$\text{Eq. 22} \quad h_{1,u} = \frac{A_{1,u}^{(\infty)}}{A_{1,u}^{(\infty)} + A_{1,a}^{(\infty)}} = \frac{1}{1 + \frac{\lambda_a}{\lambda_1 + \lambda_d + \lambda_v}}$$

Eq. 22, also derived by Jacobi (Ja72), reveals that the unattached fraction of ^{218}Po is independent of λ_p , the deposition rate constant for unattached activity.

The individual 'equilibrium' factors, g_j , above should be distinguished from the radon daughter equilibrium factor F , referring to potential alpha energy (Eq. 5 above). From a knowledge of the g_j 's, F can be calculated using the proper weighting factors (cf Eq. 4 in section 2 above).

4.2. TYPICAL ELIMINATION RATES AND EER VALUES.

Typical values of the different rate constants in Figure 1, have been published by several groups. In Table 3, the values from the review by Bruno (Br83) are given, but we have added the daughter decay constants and the deposition rate constant for attached activity.

TABLE 3.

SYMBOL	NAME	VALUE h^{-1}
0	radon decay constant	0.00755
1	^{218}Po decay constant	13.64
2	^{214}Pb decay constant	1.55
3	^{214}Bi decay constant	2.09
a	attachment rate constant	20-180
d	depos. rate const. (attached)	0.07-0.7
p	depos. rate const. (unatt.)	1-200
v	ventilation rate constant	0.1-1.25

In Table 3 the deposition rate constants for attached activities have been taken from Jacobi (Ja72), Clough (Cl73), Wicke (Wi79) and Knutsson (Kn83). A lower limit of $\lambda_v = 0.5 \text{ h}^{-1}$ is set by Bruno (Br83), however, in Sweden values of λ_v down to 0.1 h^{-1} are not uncommon, (Sæ81), and we have therefore changed the λ_v

range in Table 3. The probability of detachment, d_j , is set to zero in the room model calculations of Jacobi (Ja72), Postendörfer (Po78) and Bruno (Br83). The only recoil probability that may be of significance is a_1 . In room model calculations $a_1 = 0.5$ (Ja72, Kn83) and $a_1 = 0.83$ (Me76, Br83) have been used.

During the last decade numerous surveys of indoor radon and/or radon daughter levels have been performed throughout the world. For a guide to these measurements the reader is referred to the UNSCEAR report (UN82) and to papers included in the special radon issue of Health Physics (HP83). There exists early literature (cf Me77 for references) on measurements of 'radium emanation' indoors, but the first systematic investigation was done by Hultquist (Hu56). When only radon is measured in dwellings, most authors assume an equilibrium factor, $F = 0.5$. That this value is a reasonable choice, for low ventilation rates ($< 0.3 \text{ h}^{-1}$), has recently been proven experimentally and in model calculations (Sw83). In the interval $0.30 < \lambda_v < 0.60 \text{ h}^{-1}$, Swedjemark (Sw83) found F factors of between 0.21 and 0.66 with a mean of 0.43.

The national average of the equilibrium radon daughter concentration, (EER), including only houses on 'normal' ground, is 53 Bq m^{-3} (NRP83, Bu82), but large groups are exposed to much higher RnD concentrations. From a large track-etch detector survey of 12 000 houses, it is estimated that almost 10 % of these houses have EER-values larger than 1000 Bq m^{-3} (Hi82), but these houses were selected for measurement, because high radon daughter levels were suspected. Regions exhibiting low average radon-daughter concentrations are naturally also encountered. The average EER-value in 210 houses on the island of Öland, measured during the first half of the year 1983, was 36 Bq m^{-3} (He84). In Section 5 below the connection between EER and effective dose equivalent rates is elucidated.

5. RADON DOSIMETRY

The absorbed dose to the respiratory airways from inhalation of radon is due to radon itself and its daughters. Theoretically to arrive at an absorbed dose in different lung tissues from an air concentration of radon and its daughters, several things must be known:

- i. the activity concentration of radon and its daughters in the air,
- ii. the size distribution of the aerosol bearing the daughters, including the daughter fraction unattached to aerosol particles,
- iii. the breathing rate,
- iv. the deposition probability of the inhaled activity in different parts of the respiratory system,
- v. the elimination rate of the activity deposited or dissolved in the airways,
- vi. the thickness of the intervening tissue layer between deposited activity in the lung and the tissue of interest (the 'target' tissue).

Several of these points are interconnected, e.g. the deposition probability (iv) can not be evaluated accurately if the size distribution of the aerosol (ii) and possible changes in aerosol characteristics which may take place in the airways are not known.

It was recognized early that the epithelial cells of the bronchi were the target tissue as the carcinomas associated with radon exposure were of bronchial origin. But absorbed dose- and 'dose equivalent' calculations during the 1940's took only the inhaled radon and its subsequent decays in the lung into account. Bale (Ba51) pointed out that the absorbed dose from inhaled decay products will by far exceed the absorbed dose from radon itself. According to the rough calculations of Bale the lung dose was about three orders of magnitude larger from the daughters than from radon itself, and soon Harvey (Ha53) showed that the retention of inhaled daughters in the respiratory system was about 50 %. Taking into account the fact that the mean biological residence time of the deposited radon daughters in the mucous layer or lung alveoli was long compared with physical decay times, it was natural to consider the inhaled potential decay energy of the radon daughter as the relevant risk parameter. Furthermore, the dose equivalent from gamma- and beta decays is negligible compared with the alpha radiation, with its higher biological effectiveness. The connection between the potential alpha energy inhaled and the corresponding dose equivalent to the respiratory system was not known accurately enough, so authorities and

organisations preferred to give occupational limits expressed as potential alpha energy concentrations in air (cf Section 3 above), instead of the dose equivalent limits.

Several lung irradiation models were published during the sixties and seventies (e.g. Ja64, Al64, Ha67, Ha72), with the aim of deriving conversion factors between exposure quantities (e.g. WL or J h m^{-3}) and the absorbed dose to different parts of the lung tree. The interest in radon daughter dosimetry at this time was aroused from the findings of high RnD concentrations in uranium mines in the USA during the 1940's and 50's. The median radon concentration in 36 uranium mines on the Colorado plateau was 700 kBq m^{-3} (Ba51).

The in-depth evaluation of all the parameters possibly influencing the dose to the respiratory organs revealed, among other things, the importance of the unattached fraction of the short-lived daughters. The high deposition probabilities of the unattached daughters lead to dose-exposure factors about ten times higher than for attached activity. Jacobi (Ja77) found conversion factors between the dose to the bronchial tissue and the potential alpha energy exposure ranging from 0.007 to $0.4 \text{ mGy/J h m}^{-3}$ in his survey of different models. But he noticed that the major part of the discrepancies was due to different assumptions concerning unattached fractions and aerosol size distributions, made by the various authors. Correcting for the differing aerosol characteristics, Jacobi narrowed the range of conversion factor applicable to mining down to 0.01-0.04 mGy/J h m^{-3} . The figures given refer to one of the target tissues in the lung, the basal cells of the bronchi. The alpha-particle range in tissue is of the same order as that in the mucous layer covering the basal cells; therefore a critical parameter in every lung-dose model is the mucous layer thickness (Ja83).

A renewed interest in lung-dose model calculations followed the concern about radon daughter irradiation in dwellings. The mine models have to be modified as the aerosol conditions and the breathing rates are different in a dwelling. The fact that children are exposed has also to be considered. Age-dependent conversion factors have been derived by Hofmann (Ho80). The results of the extensive studies of radon daughter lung dosimetry by Jacobi and Eisfeld (Ja80, Ja81) and James et al (Jam80) have been compared by an OECD/NEA expert group (Jam81). The International Commission on Radiological Protection (ICRP) refers to the lung models of Jacobi-Eisfeld (J-E), James-Birchall (J-B) and ICRP when deriving dosimetrically based occupational limits (ICRP81). Recently James (Ja83) presented a comprehensive comparison of the three models by Jacobi, James and Harley-Pasternak (Ha81).

From a study of the modern realistic lung-dose analysis it can be concluded that

- i. the conversion factor between an absorbed dose and potential alpha energy is strongly dependent on the fraction of unattached radon daughter activity,
- ii. two different target tissues, the bronchial basal cells and the cells in the pulmonary region should be considered separately,
- iii. the identification of the most risk-relevant regions within these targets are uncertain (Ho83).

From i. it follows that the potential alpha energy concentration is not an appropriate measure of the risk in conditions where the unattached fraction varies. But the question is what to use instead. For want of better knowledge on how the deposited radiation energy in the respiratory tract should be weighted for the best risk assessment, we have used the calculation principles of the ICRP (ICRP81) in paper III to evaluate the impact of air cleaning. Exploiting the regional lung dose-concept of ICRP publication 32 (ICRP81), with equal weighting factors ($w_T = 0.06$) for both the bronchial basal cells and the pulmonary region, yields conversion factors, $\dot{H}_\alpha / \dot{I}_{\text{pot}}$, as a function of the fraction of unattached potential alpha energy, f_p ,

$$\text{Eq. } \dot{H}_\alpha / \dot{I}_{\text{pot}} = \underbrace{(18+170f_p)}_{\text{basal cells}} \cdot 0.06 + \underbrace{5.2(1-f_p)}_{\text{pulm. region}} \cdot 0.06 = 1.39 + 9.9f_p \quad (\text{Sv J}^{-1})$$

23a

(J-E model)

$$\text{Eq. } \dot{H}_\alpha / \dot{I}_{\text{pot}} = \underbrace{(14+560f_p)}_{\text{basal cells}} \cdot 0.06 + \underbrace{2 \cdot 0.06}_{\text{pulm. reg.}} = 0.96 + 34f_p \quad (\text{Sv J}^{-1})$$

23b

(J-B model)

$\dot{H}_\alpha$ is the effective dose equivalent rate and $\dot{I}_{\text{pot}}$ is the potential alpha energy inhalation rate (J h^{-1}). If the mean breathing rate is denoted b ($\text{m}^3 \text{h}^{-1}$), then $\dot{I}_{\text{pot}}$ and the air concentration of potential alpha energy C_{pot} (J m^{-3}), are simply connected:

$$\text{Eq. 24} \quad \dot{I}_{\text{pot}} = b C_{\text{pot}}$$

$$C_{\text{pot}} (\text{J m}^{-3}) = 4.80 \cdot 10^4 C_{\text{pot}} (\text{WL}) = 1.80 \cdot 10^8 \text{ EER} (\text{Bq m}^{-3})$$

Eq. 23 assumes an activity median diameter of about $0.25 \mu\text{m}$ (ICRP81), and the quality factor for alpha particles $Q = 20$. The presumed diffusion coefficient for the unattached activity is $0.05 \text{ cm}^2 \text{ s}^{-1}$.

In Eq. 23 the dose equivalent to the basal cells dominates, especially at high values of the unattached fractions, f_p . The identical weighting factor 0.06 for both basal cells and the pulmonary region is questionable, seen from a radon epidemiological stand point. As all radon daughter induced tumours are localized in the upper bronchi, it would seem reasonable to put more, if not all, (0.12) weight on the bronchial dose equivalent (Ja83). In paper II, however, we have strictly followed the recommendation of the ICRP in putting $w_T = 0.06$ for both target regions.

Eq. 23 can be written in terms of EER instead of the intake of potential alpha energy. Assuming, as in paper III, a mean breathing rate of $1.2 \text{ m}^3 \text{ h}^{-1}$ we have

$$\begin{aligned} \text{Eq. 25a} \quad \dot{H}_\alpha / \text{EER} &= \overbrace{7.2(1-f_p) + 75.2f_p}^{\text{basal cells}} + \overbrace{2.08(1-f_p)}^{\text{pulm. region}} = \\ &= 9.3(1-f_p) + 75.2f_p \quad \left(\frac{\mu\text{Sv h}^{-1}}{\text{kBq m}^{-3}} \right) \quad (\text{J-E model}) \end{aligned}$$

$$\begin{aligned} \text{Eq. 25b} \quad \dot{H}_\alpha / \text{EER} &= \overbrace{5.6(1-f_p) + 230f_p}^{\text{basal cells}} + \overbrace{0.8}^{\text{pulm. reg.}} = \\ &= 6.4(1-f_p) + 230f_p \quad \left(\frac{\mu\text{Sv h}^{-1}}{\text{kBq m}^{-3}} \right) \quad (\text{J-B model}) \end{aligned}$$

The formula for calculating the equilibrium equivalent radon concentration, EER, is given by Eq. 4 above. The mean breathing rate of $1.2 \text{ m}^3 \text{ h}^{-1}$ in ICRP 32 (ICRP81) is defined for occupational exposure. For domestic situations the lower breathing rate of $0.75 \text{ m}^3 \text{ h}^{-1}$ is recommended (UN82). Equations 25a and 25b are

linearly dependent on breathing rate and for $b = 0.75 \text{ m}^3 \text{ h}^{-1}$ Eq. 25a and Eq. 25b become:

$$\begin{aligned} \text{Eq. 26a} \quad \dot{H}_\alpha / \text{EER} &= 4.5(1-f_p) + 47f_p + 1.3(1-f_p) \\ &= 5.8(1-f_p) + 47f_p \quad \left(\frac{\mu\text{Sv h}^{-1}}{\text{kBq m}^{-3}} \right) \quad (\text{J-E model}) \end{aligned}$$

$$\begin{aligned} \text{Eq. 26b} \quad \dot{H}_\alpha / \text{EER} &= 3.5(1-f_p) + 144f_p + 0.8 \\ &= 4.3(1-f_p) + 145f_p \quad \left(\frac{\mu\text{Sv h}^{-1}}{\text{kBq m}^{-3}} \right) \quad (\text{J-B model}) \end{aligned}$$

The strong dependence of the effective dose equivalent rate on the unattached fraction, f_p , is obvious from the formulae above. In the J-E dosimetric model the unattached and attached fractions are of equal importance when f_p is about 14 %. The corresponding figure for the J-B model is only 3 %. The reason for the smaller weighting factor for the unattached activity in the J-E model is traced back to the high clearance rate of the unattached activity deposited in the bronchi, due to an assumed rapid uptake ($T_{1/2} = 15 \text{ min}$) into the blood (Ja83). Equations 26a and 26b can be utilized to estimate the effective dose equivalent due to radon daughters. From the reference value of $f_p = 0.025$ in homes (Ja83) we have from Eq. 26 $\dot{H}_\alpha / \text{EER} = 6.8_3$ and $7.8_2 \mu\text{Sv per (kBq h m}^{-3})$ from the J-E and J-B dose models respectively. If we further assume an indoor occupancy factor of 0.8, $f_p = 0.025$ for outdoor air and use the mean indoor and outdoor EER values 5_3 and 2 Bq m^{-3} , in Swedish homes (NRP83) on natural ground, we arrive at $\dot{H}_\alpha = 2.5_3 + 0.0_3 = 2.5_6 \text{ mSv a}^{-1}$ (indoor + outdoor) in the J-E model. The corresponding J-B values are about 10 % higher. In the calculation the mean breathing rate $b = 1.0 \text{ m}^3 \text{ h}^{-1}$ has been applied outdoors (UN82). The conversion values used here (cf Eq. 23 with $f_p = 0.025$) are slightly smaller than the reference value of 2 Sv per J inhaled potential alpha energy, chosen in the UNSCEAR report (UN82). The average effective dose equivalent rate from external gamma radiation in Sweden is 0.6 mSv a^{-1} (NRP83); excluding the contribution from cosmic radiation, about 0.3 mSv a^{-1} . In several countries the average national EER is estimated to be in the range $15 - 25 \text{ Bq m}^{-3}$ (UN82, Table 40). In those countries the indoor radon daughter exposure constitutes the major part of the 'natural radiation background', expressed in effective dose equivalent.

In the time prior to the existence of reliable dose models and the publication of ICRP report 26 (ICRP77a), defining an effective dose equivalent, only little

attention was paid to exposure from radon daughters in dwellings. It is relevant to talk about an era before and after IRCP 26. The effective dose equivalent quantity was a means, though inexact, putting all partial body irradiations on a common basis, from the point of view of carcinogenic risk. In older listings of background radiation levels the lung dose from radon daughters is not mentioned, or if it is, the given values are difficult to compare with e.g. whole body gamma irradiation. The UNSCEAR reports illustrate the growing awareness of radon as a significant background source. In the 1962 UNSCEAR (UN62) report the increased external gamma radiation dose rate in buildings constructed from lightweight concrete based on alum shale are listed (Hultquist's measurements, Hu56), but the topic of indoor radon and thoron is only touched upon 'concentrations in confined volumes such as buildings will be considerably higher if ventilation is poor or absent'. The UNSCEAR report from 1966 has much more to say. Between 1962 and 1966 two dose models (Al64, Ja64) calculating the bronchial- and lung tissue dose were published, and several paragraphs (roughly half a page) in UNSCEAR 66 deal specifically with indoor exposure to radon- and thoron decay products. Lung doses were still not included in the summarizing table (Table XVII), covering dose rates from natural sources, but paragraph 149 contains the following comment; 'Dose rates to the lung tissues are not given in the table since no exact estimate is available, but those to the epithelium of segmental and lobar bronchi are believed to be of the order of several hundreds of millirads per year.' (1 millirad= 0.01 milligray). In the 1972 UNSCEAR report (UN72), Table 20, (p.83) summarizes the dose rates from natural sources in 'normal' areas. Dose rates to gonads, bone-lining cells and bone marrow from ^{222}Rn are given, but no lung doses. But paragraph 146 comments on Table 20; 'In addition to the doses included in table 20, yearly doses in the range 50- 200 millirads are received by the basal epithelial cells of the segmental bronchi from inhaled radon daughters. Their actual value are still uncertain owing to the many assumptions that underly the estimates.' Later UNSCEAR reports (UN77, UN82) treat radon and thoron and their decay products more extensively. In the 1982 report text and tables in Annex D 'Exposures to radon and thoron and their decay products' take up no less than sixty pages, excluding the nine pages listing references.

6. MEASUREMENT TECHNIQUES

In this section a brief description of the different measuring principles and techniques for radon and radon daughters is given. Only methods utilized in papers I - IV are treated, with a particular emphasis on methods of fundamental importance for the conclusions drawn. The reader having a general interest in radon- and radon daughter measurement techniques, is referred to several review papers published, e.g. by Budnitz (Bu74), George (Ge80) and Breslin (Br80).

Only methods involving first a collection of activity and then a subsequent measurement of the activity are relevant to this investigation. If the radio-activity concentration is low, the activity is collected via accumulation on some kind of filter. The measurements may be based on different physical principles, e.g. gas ionization in an ionization chamber. The detection methods used in this investigation are limited to alpha scintillations from zinc sulphide (ZnS) and alpha spectrometry using solid state detectors.

6.1. RADON SAMPLING AND MEASUREMENT

The established scintillation flask, also called the Lucas flask, is used as the detector for radon throughout papers I to IV. The technique of using zinc sulphide (ZnS) as a detector for alpha radiation is very old, and was used by Crooke in his 'spinhariscope', (Cr03). In this old application of ZnS, the light flashes were counted by eye. In modern designs the inside surfaces of the scintillation flask are coated with silver-activated zinc sulphide. The bottom of the flask is made from a light-transparent material and the flask are equipped with one or two valves. Placing the scintillation flask on a photomultiplier (PM) tube, the alpha scintillations can be counted by ordinary pulse electronics. The scintillation flask got its alternative name from the improved detector design published by Lucas (Lu57).

In this investigation grab samples are taken in single valved preevacuated scintillation flasks, either of commercial (LAC-3, Johnston Lab, Maryland, USA) or our own design. In the latter case the phosphor, obtained from Radium Corporation, Hackettstown, New Jersey, USA, is sprayed onto the inside of cheap water-seal cups, with a small pressurized air sprayer. The air in the grab samples taken is always prefiltered, to prevent dust and radon daughters from entering the flask. The background count rates of the flasks are individually determined and vary between about 0.01 and 2 counts per minute, depending on the

type and the radon exposure history of the flask, as well as the working voltage of the PM tube.

By permanently mounting the scintillation flask in the PM-tube housing continuous radon detector is possible. A double-valved through-flow scintillation flask detection is the subject of paper II. As in the case of grab sampling the inlet air is filtered to remove radon daughters. Whilst inside the detector some radon atoms will decay, and a fraction of the daughters born, will be deposited on the detector surfaces, while the others are swept out by the air stream. The deposited daughters give the continuous flask detector a 'memory' of prior radon concentrations. The intended use of a continuous radon detector is at places where the radon concentration in air varies, and naturally the radon daughter memory effect of the through-flow detector is a drawback. The first continuous flask monitor was constructed at the former HASL by Harris et al (Ha57). Two decades later at the same laboratory, (Environmental Measurement Laboratory, Dept. of Energy, New York), a procedure was developed to correct for the memory effect (Co77, Th79). Using the method described in paper II, the true radon concentration C_n in time interval n , can be written

$$\text{Eq. 27} \quad C_n = \epsilon^{-1}(S_n + m_1 S_{n-1} + m_2 S_{n-2} + \dots m_i S_{n-i} + \dots)$$

where S_n is the accumulated number of counts (corrected for back-ground) in the time interval n , and m_i are the memory coefficients. The total counting efficiency, ϵ (counts per Bq m^{-3}), is composed of a counting efficiency, for radon, ϵ_r , and a counting efficiency for radon daughters, ϵ_d . The calibration procedure to evaluate the coefficients m_i is given in paper II. As the coefficients m_i are directly connected to the elimination coefficients in the flask and the elimination rate is flow-dependent, the m_i coefficients are also flow-dependent. However, the dependence of the m_i 's on flow rate is small, in the flow-rate interval used in paper II. The reason is that the plate-out process is only slightly influenced by the air-flow changes in question.

The scintillation flask is limited to a volume of about two litres or less. The restriction in volume is imposed by both the alpha particle range in the flask and the difficulties in obtaining efficient light-collection from large-volume flasks. The net pulse rate from a one-litre scintillation flask is about 0.1 counts per minute at a concentration of 1 Bq m^{-3} , if radon equilibrium in the flask is assumed. The detection limit for radon will be different depending on the background count rate of the flask and the counting time for the sample. From the figures given it can be concluded that the lower limit of detection, taking grab samples, is in the interval 1-10 Bq m^{-3} , or even higher if a small

flask has a high background count rate.

To increase the sensitivity in grab-sample measurements, the radon in air can be concentrated to a small volume either by condensation, adsorption or chemical processes. In order to manage the extremely low radon concentrations met in paper IV, adsorption of radon on cooled charcoal was utilized. The knowledge that charcoal is an efficient adsorbent of radon is old, described by Rutherford in 1906 (Ru06) and Boyle in 1907 (Bo07). Several authors have investigated the dynamics of charcoal traps for inert gases, e.g. (St55, St81, Bo81, and Sc80). One method of analysing the adsorption behaviour of charcoal traps is the equilibrium compartment model. In this model the trap is treated as a series of N compartments of equal volume. Exposing the trap for a constant volume flow of radon-free air, v , and a short radon pulse at time $t=0$, the breakthrough volume of air V is approximately related to the dynamic adsorption coefficient k by the expression (Bo82)

$$\text{Eq. 28} \quad V = t_b k m / t_m$$

where m is the mass of the charcoal and t_b and t_m are the times of breakthrough and maximum activity in the outlet air, respectively. The breakthrough volume V is equal to $v t_b$. From the radon elution curve the number of compartments, N , can be calculated (St81).

$$\text{Eq. 29} \quad (t_m / \Delta t)^2 = 0.180 N - 0.194$$

In equation 29 Δt is the full width at half maximum concentration. The dynamic adsorption coefficient k is strongly dependent on temperature. The temperature dependence can be written (Bo78)

$$k = s_1 \exp(s_2/T)$$

Where s_1 are positive constants and T ($^{\circ}\text{K}$) is the charcoal temperature. At $T = 273^{\circ}\text{K}$ the dynamic adsorption coefficient is about three times higher than at 293°K , as reported by Scheibel et al (Sc80).

6.2. RADON DAUGHTER PRODUCTS

Originally, the short-lived daughter activities in air were often measured directly using ionization- or scintillation methods. But at low concentrations, in outdoor air for instance, some sort of accumulation was necessary. For this

purpose electric-field methods, e.g. high voltage wires and air flow electric condensers, were frequently utilized. In response to a paper by Bale (Ba51), stressing the dominating role played by radon daughters in inhaling radon-laden air, several 'mechanical' filter methods were born. The concentration in air of short-lived daughters could be deduced from an air sample drawn through a filter of fine porosity, followed by an alpha count of the filter. The first sampling and measuring regimes were based on gross alpha measurements in one (Ku56, Ro72) or several (Ts53) time intervals subsequent to the air sampling. A determination of the individual air concentrations of all three short-lived daughters is possible with the three-interval regimes, like Tsivoglou's (Ts53). In later developments alpha counting during air sampling (e.g. Cl78a, Cl78b) increased the statistical precision of the ^{218}Po determination. Increased sensitivity and precision may also be obtained if the alpha-decay count rate of the filter is measured continuously after sampling. The measuring time can be chosen to be up to an hour or more. The individual daughter concentration in air is derived by means of a least-squares fit to the decay curve (Ra69). The advantage of the gross alpha methods is that large-area alpha detectors are available. High-volume samples, using filters and detectors (e.g. ZnS) of areas of a few square decimetres, make the gross alpha measurements very sensitive. The methods are easy to use in the field, and since long measuring regimes such as Kusnetz' (Ku56) are frequently used in mines.

Using gross alpha three-interval regimes (Tsivoglou and modifications, of IAEA76), it is difficult to achieve enough precision in the determination of the short-lived ^{218}Po ($T_{1/2} = 3.05$ minutes). As mentioned the precision may be improved by counting during sampling, but then the air-sampling head, must be of a more complex design. The detector must be situated in front of the filter. Significant losses of unattached activity can not be excluded in such a geometry.

A direct determination of the first daughter ^{218}Po is possible using alpha spectrometry. The 6.00 MeV ^{218}Po peak is easily resolved from the 7.69 MeV peak of ^{214}Po and influences from other alpha-decaying nuclides can be corrected for. Figure 2 shows an example of an alpha spectrum containing peaks from decay products of three natural decay series. The alpha spectrometry method for air-borne radon daughters was proposed by Martz et al (Ma69). For an unambiguous determination of the individual radon daughter concentration two counting time intervals are necessary. From the net accumulated alpha pulses from ^{218}Po (P_1) and ^{214}Po (P_2) in the first interval and from ^{214}Po (P_3) in the second interval the air concentration C_i of the i :th daughter can be deduced:

counts per channel
a.u.

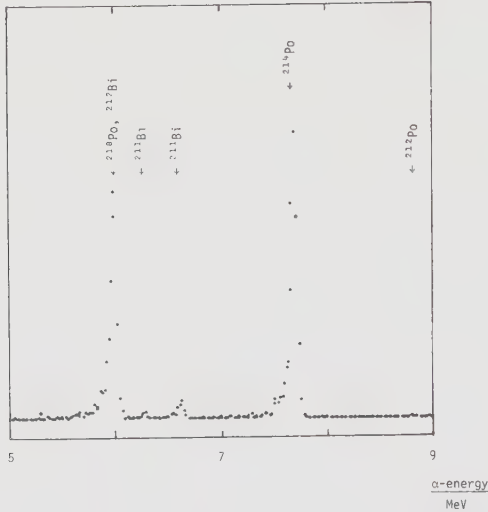


Figure 2. An alpha spectrum obtained from the radon room with uranium ore as a radon source, (Millipore FS filter).

$$C_1 = k_1 P_1 (1/v\epsilon) \quad (\text{Bq m}^{-3})$$

$$\text{Eq. 30} \quad C_2 = (k_2 P_1 + k_3 P_2 + k_4 P_3)(1/v\epsilon) \quad (\text{Bq m}^{-3})$$

$$C_3 = (k_5 P_1 + k_6 P_2 + k_7 P_3)(1/v\epsilon) \quad (\text{Bq m}^{-3})$$

where ϵ and v are the alpha counting efficiency of the detector and the air volume sampling rate respectively. The coefficients k_i are dependent on the sampling time and the subsequent counting regime. In the first step our computer program (Pe81) calculates the coefficient values for an arbitrary choice of sampling time and length of the two counting time intervals. And secondly, the concentration values C_i are calculated, following equation 30.

The rationale of computerized alpha-spectrometry evaluation, is the freedom to introduce various sampling- and counting regimes without tedious recalculation of coefficients. Due to the short half-life of ^{218}Po , the timelag, t_1 , to the first counting interval should be kept to a minimum. The accuracy of the alpha-spectrometry method has been discussed by Martz et al (Ma69) and Jonassen and Hayes (Jo74). In the investigation in paper III, alpha spectrometry was used throughout, for measurements on both filters and wire screens, except for the investigations reported in Figure 8 III.

One of the problems met in paper III was how to separate the attached and unattached radon daughter activities. Upon consulting the literature several methods were found to be available. The wire screen technique (Th72) was chosen, as this technique is easy to adapt to alpha spectrometry measurements, a direct measurement of the unattached activity is possible and commercial filter holders can be used with little or no modification. The wire screen can be considered as an array of short diffusion tubes in parallel. When drawing air through the wire screen, there is a finite probability for particles to be deposited on the wires either by inertial impaction, molecular or eddy diffusion. In laminar flows of small particles, say less than one micrometre, only molecular diffusion is of importance. Then the highly diffusible unattached radon daughters have a large probability of becoming attached to the wires. The aerosol particles, much less mobile across the air flow, can pass the wire screen almost unaffected.

Even if the wire screens, as passive devices, are simple to use, careful investigations of collection and counting efficiencies must precede usage. In paper III the collection efficiency of the wire screen has been both experimentally and theoretically analysed. Two theories have been applied, the semi-empirical formula suggested by Thomas (Th72) and the theoretical efficiency equation based on the fan model of filtration theory published by Yeh, Cheng and Orman (Ye82). Thomas' formula for the collection efficiency, u , can be expressed as a function of the dimensionless parameter h ,

$$\text{Eq. 31} \quad u = 1 - 0.82 \exp(-23.3h) - 0.18 \exp(-1670h)$$

where $h = D\varphi/v(\varphi+y)^2$
 φ = screen wire diameter
 y = aperture of the wire screen
 D = diffusion coefficient
 v = laminar air velocity

As pointed out by the authors (Th72), the semi-empirical approach behind Equation 31 cannot be expected to give accurate collection efficiencies for screens and flow rates differing greatly from those of their study ($(\varphi+y)^{-1}$ from 1.6 to 32 cm⁻¹). The flow rate interval covered by Thomas et al (Th72) is 5 - 50 cm s⁻¹.

Equation 31 is only applicable to unattached particles. In order to evaluate theoretically the collection efficiency of the wire screen for aerosol particles, the diffusion tube equation can be used in the limit of small h (So79), as Vooren et al (Vo82) have done. We, however, have instead exploited the formalism

developed by Cheng and collaborators (Ch80). Originally derived for wire screen diffusion batteries, the collection efficiency, a , of a single wire screen, can be written as a function of the Pechlet number $P = v(\phi/D)$ and the interception parameter $R = \phi/\varphi$ (ϕ = particle diameter):

$$\text{Eq. 32a} \quad a = 1 - \exp - (b_0 P^{-2/3} + b_1 R^2 + b_2 P^{-1/2} R^{2/3})$$

where the coefficients b_i are solely dependent on the wire screen geometry. The b_i 's can be written in terms of the parameters B and w , which are dependent on the solid volume fraction, α , of the screen and the screen thickness, t , (Ye 82):

$$\begin{aligned} b_0 &= 2.7 B / \ln 10 \\ b_1 &= 2.0 B / (2 w \ln 10) \\ b_2 &= 1.24 B / (w^{1/2} \ln 10) \\ B &= 4\alpha t / \Pi (1-\alpha) \varphi \\ w &= -0.5 \ln \frac{2\alpha}{\Pi} + \frac{2\alpha}{\Pi} - 0.75 - 0.25 \left(\frac{2\alpha}{\Pi} \right)^2 \end{aligned}$$

For an aperture $y = 0.25$ mm and the wire diameter $\varphi = 0.16$ mm (the screen used in paper III) the $b_1 R^2$ term in Eq. 32a can be neglected. If we approximate the solid volume fraction to $\alpha = 4(\varphi + y) / \Pi \varphi$ and the screen thickness to $t = 2\varphi$, we have

$$\text{Eq. 32b} \quad a = 1 - \exp\{- (1.17 P^{-2/3} + 7.97 P^{-1/2} R^{2/3})\}$$

Relating the particle diameter, ϕ , and the diffusion coefficient D (Ye82) by

$$D = kT\beta / 3 \Pi \mu \phi$$

where k is the Boltzmann constant, T is the absolute temperature, β is the Cunningham slip correction, and μ is the air viscosity, the collection efficiency, a , has been calculated. In paper III the collection efficiency, a , is displayed for two air-sampling velocities, as a function of particle diameter and for log-normal particle size distributions.

To experimentally evaluate the collection efficiency of the wire screen, three screens, in series, in front of a back-up filter, have been used. Based on assumptions given in paper III, the collection efficiencies for attached activity (a_i) and unattached activity (u_i) for the i :th daughter can be derived. The

formulae are given in paper III. Due to the low absolute values of the efficiency a , of the order of 0.01, the experimental accuracy of a_i is low. On the whole, the theoretical and experimental results of the wire screen collection efficiencies for both attached and unattached particles are consistent (cf Figure 3 in paper III) as long as laminar air flow prevails.

Once the a_i 's and u_i 's have been determined, a single wire screen in front of an absolute filter, suffice as an air-sampling set-up, to separate attached (C_a) and unattached (C_u) activity concentrations.

$$C_u = \{S - a(S + F)\} / (u - a) \quad (\text{Bq m}^{-3})$$

Eq. 33

$$C_a = (S + F) - C_u \quad (\text{Bq m}^{-3})$$

where

S = the activity concentration in air corresponding to the activity on the wire screen.

F = the activity concentration in air corresponding to the activity measured on the back-up filter.

u = the collection efficiency of the wire screen for unattached activity.

a = the collection efficiency of the wire screen for attached activity.

In order to determine S and F in Eq. 33, the counting efficiency of the surface-barrier detector for alpha activity on wire screens and on filters must be known. In paper III the counting efficiency for the wire screen relative to the filter is of primary importance. (The absolute values are less essential in paper III, where relative values are sufficient to evaluate the effect of air cleaning). To relate alpha-counting efficiencies, the gamma radiation from the short-lived daughters on screens and filters has acted as reference radiation. The procedure is based on the assumption that both wire screens and filters are thin plane gamma sources, with negligible self-attenuation. By inserting a small diffused-junction alpha detector into a NaI well-crystal gamma detector, alpha- and gamma radiation can be detected simultaneously (paper III). Another possibility is to count for several consecutive periods, of say a few minutes, alternately with a surface-barrier detector and with a NaI crystal. It is in this case convenient to utilize the time period 10 -20 minutes after sampling, when the ^{214}Bi gamma- and ^{214}Po alpha intensities are approximately constant with time. Furthermore, the gamma radiation from ^{214}Bi and the ^{214}Po alpha radiation,

can be considered as coincident in this application, due to the short half-life of ^{214}Po . These alpha- and gamma measurements reveal that the alpha counting efficiency for wire screens is dependent on air velocity in the interval $u < 10 \text{ cm s}^{-1}$, supporting earlier findings by James et al (Jam72). At sampling velocities above 10 cm s^{-1} the alpha counting efficiency for a wire screen is twenty percent lower than for a filter.

The technique of sampling radon decay products on filters is applicable to long-lived daughters as well. The relatively small air samples discussed (50 m^3) in paper IV, prohibit a direct measurement on the filters, as the air concentration of long-lived daughters is very low in the arctic. For assessment a relatively simple chemical separation technique is chosen. Easily soluble filters are used and from the dissolved filter solution ^{210}Po is deposited onto nickel plates. The ^{210}Po concentration is measured with surface-barrier detectors having very low background. Counting times from several days up to two weeks are still needed to achieve acceptable counting statistics.

As the long-lived daughters have been collected through a large pipe and, in addition, the air sampling head is of the non-open type, activity loss must be evaluated. This has been done in paper IV, with the result that no significant loss of long-lived activity could be detected. These findings are reasonable, as the aerosol carrying the long-lived daughters is well aged, and therefore the very small particles have been removed by coagulation processes.

6.3. EXHALATION MEASUREMENTS VIA THE ACCUMULATION METHODS

In the recent survey of radon transport phenomena (Co81) several exhalation methodologies are reviewed. All direct methods of measuring radon exhalation involve a vessel of some kind to accumulate the exhaled radon. On surfaces, the vessel, with at least one end open, is placed over the surface and may be sealed. Small samples can also be placed inside canisters. From the growth of radon inside the vessel or canister after closing, the radon exhalation is deduced. Special forms of the accumulation method are the flow- and the adsorption methods. In the flow method (Pe65) the accumulating vessel is connected in a closed loop to a through-flow radon trap. The air flow in the vessel should, as nearly as possible, simulate conditions met outdoors on the ground. The adsorption method implies filling the accumulating vessel with charcoal. On surfaces the vessel may also be open towards the room. This technique introduces fewer disturbances than a closed container. In the following description the

discussion will be limited to air-filled accumulating vessels used for exhalation determination of enclosed samples, even if some conclusions may also be relevant for the adsorption method.

To assess the exhalation from the enclosed sample the radon concentration in the vessel must be measured. This can be done by taking one or several grab samples of air from the vessel. If a single sample is taken after a certain accumulation time, a constant exhalation rate from the enclosed sample is assumed. This exhalation rate is easily calculated when the free volume in the vessel is known. The radon concentration measured should be corrected for radioactive decay and leakage of the vessel if the accumulation time is long. Long accumulation periods, several days as used e.g. by Zupancic (Zu34) and Lindmark (Li84), combined with single grab samples cannot be recommended, as the exhalation rate does not remain constant. Applying the accumulator method to outdoor ground surfaces, Clements and Wilkening (Cle73) recommend that the accumulation time should be less than one or two hours.

To avoid the uncertainty inherent in the single-sampling method, the initial growth of the radon concentration can be traced by repeatedly taking small samples of air from the vessel. Due to the extensive work of Jonassen (Jo80, Jo83) this method is now in common use. The initial radon concentration growth in the vessel is used to assess the free exhalation rate of the enclosed sample. By free exhalation is meant the exhalation rate of the sample during equilibrium conditions in free space. The grab samples taken dilute the air in the vessel and introduce periodic reductions in pressure. The total volume of the grab samples should preferably be small compared with the free air volume of the vessel, to avoid significant problems of dilution and pressure change. Working with perfectly air tight enclosures, the pressure drop can be avoided by deliberately replacing the grab air via a second valve in the vessel. Care must be taken that the grab-sample air is representative of the air in the vessel, and not, for example, diluted by the replacement air. The outer-volume air in the vessel should be stirred to avoid radon concentration gradients, if the height of this outer volume is large. With carefully designed experimental conditions, disturbing effects of temperature or pressure changes, dilution, etc. can be avoided. During controlled conditions the only transport mechanism causing exhalation is diffusion. Such ideal conditions are the subject of paper I. The implications of the time-dependent diffusion theory on exhalation measurements is elaborated on in section 7 below.

7. RESULTS AND DISCUSSION.

The aim of this section is to summarize and discuss the findings in papers I-IV. Only the more important implications and results are dealt with here. Details are found in the respective papers. Included in this section are also some complementary experimental results obtained after the printing of papers I-IV. The first part below describes the calibration procedures common to all four papers.

7.1 CALIBRATION AND TRACEABILITY.

A standard ^{226}Ra solution (Amersham RAY.24, Gr. Britain) is used in a vacuum-tight radon emanator system, to calibrate the ZnS flasks. The ^{226}Ra solution is measured by the manufacturer in an ionization chamber, calibrated using a ^{226}Ra standard from the National Bureau of Standards, Washington D.C., USA. The overall uncertainty in the ^{226}Ra concentration as defined by Amersham is $\pm 3.7\%$. The solution is diluted in two steps before the transfer to the emanator vessel. For dilution a micropipette of nominal volume 1 ml, but calibrated to 0.964 ± 0.011 ml (± 1 s.d.) is used. The design and use of the emanator follow essentially the recommendations given by Rushing et al (Ru64). The total reproducibility using the emanator as a standard over longer periods of several months is about 5% (1 s.d.). Counting statistics contribute insignificantly to this figure.

The radiation from the short-lived radon daughters is, with a few exceptions, measured using a surface-barrier detector, calibrated using commercial alpha sources, plated on metal discs. As stressed in paper III, one should not consider such a procedure absolute, mainly due to the larger and unknown energy degradation of alpha particles in the filter source, compared with the standard. As the counting efficiency of the filters is based on plated sources, it is an upper limit. As the wire screens are calibrated relative to the filters, the same degree of overestimation applies to them. The daughter concentrations presented in paper III should be considered as relative. From preliminary absolute calibration procedures now underway, it is estimated that the RnD values given in paper III are too low by about 10-15 %. All other short-lived radon daughter detectors (surface-barrier or zinc sulphide type) utilized for measuring the activity on filters and wire screens have been calibrated relative to the reference surface-barrier detector.

The long-lived daughter concentrations are deduced by counting the alpha particles from ^{210}Po , deposited from the filter solution onto nickel discs. The sur-

face-barrier detectors (BA-19, 300 mm², Ortec, Tennessee, USA) are calibrated by means of standard alpha sources of total uncertainty about $\pm 1\%$. In this case both the samples to be measured and the calibrating sources are thin plated sources of the same diameter. The long-lived daughter concentrations given in paper IV are therefore the absolute ones, directly comparable to the radon values.

In connection to the calibration discussion above it is worth stressing the importance of reproducing exactly the distance between source and detector. For the typical geometries used (source-detector distance 4 mm, source and detector diameter of about 30 mm) when alpha-counting the filters, the counting efficiency changes by about 7 % per mm.

Air pumps of different designs and capacities have been used. The air sampled for long-lived daughters benefits from a pump equipped with a constant flow regulator (RAS-1, Eberline, Santa Fe, New Mexico, USA). All pumps have been used connected to gas volume meters. Tests between the different instruments utilized to determine air-sample volume show consistent results (the factory calibration figures have been assumed to be correct). Air density corrections due to different atmospheric pressures have not been applied.

7.2 RADON EXHALATION (paper I).

The objective of paper I is to describe the consequences of diffusion theory for exhalation measurements using the closed-can (accumulation) method. The treatment is limited to one-dimensional samples in air-tight vessels.

One important result in paper I is the proof that a sample filling up the bottom of a straight-walled vessel to a height d , exhibits the same areal exhalation as a semi-infinite sample of thickness $2d$, (Figure 1, I). A one-dimensional analysis of the sample is therefore perfectly adequate as long as the walls and bottom of the vessel are impermeable and non-absorbant for radon.

The most important issue in paper I is however, the increased insight into the exhalation phenomenon gained from studying the solution to the time-dependent diffusion equation. It is however, not necessary to solve the complicated time-dependent diffusion equation in order to reject the old interpretation of 'back-diffusion', as the following intuitive discussion will illustrate.

If we, at time zero, have the sample in the open container in equilibrium, the concentration gradient in the pores has the general trend of the curve for $t=0$, in Figure 2a I. (For simplicity we assume that the outer volume concentration of radon is nil at time zero). Closing the exhalation vessel at $t=0$, will cause the outer volume concentration to increase linearly with time. But the most superficially situated pores in the sample will follow the same rate of increase in concentration, as the well mixed outer volume does. Deeper situated pores in the sample will 'respond' to the sudden increase in outer volume concentration somewhat more slowly, due to the finite times of radon transportation involved. Thus, the immediate answer to a linear increase in outer volume concentration is a decreased concentration gradient in the surface region of the sample. Consulting Fick's first law of diffusion, we find that this is equivalent to a lowered exhalation rate. In conclusion then, the immediate response to closing the container is a depressed exhalation rate. The rate of change in exhalation rate is at a maximum initially. (How fast the change is, is determined by the outer to inner volume ratio, α , and the diffusion length, L , of the sample material). From this qualitative description it is also clear that the decreasing exhalation rate, immediately after the closing of the vessel containing the sample, has nothing to do with the high radon concentration in the outer volume of the vessel. In fact, the depression in exhalation rate is most rapid at $t=0$, when the concentration in the outer volume is zero. One important question is now: for how long after closure is the radon concentration growth in the outer volume linear with time? Already the intuitive discussion above indicates that this period of linearity may actually be zero. That it is theoretically so, is confirmed by the time-dependent solution of the diffusion equation (Eq. 9) above). Thus, the diffusion theory implies that there is no period of strict linear growth of radon in the container volume, corresponding to free exhalation. Two questions related to each other naturally arise. Firstly, how is it that radon experimentalists so often obtain apparently linear increases of radon concentration in the container outer volume? (sometimes over a day or more), and secondly, what circumstances can 'save' old 'free exhalation' results, obtained by the closed can method? The first question is easy to answer. For certain configurations of sample and measuring vessels, the reshaping time (cf definition in paper I) is so small, that a practically linear growth in radon concentration in the outer volume is measured, a growth rate corresponding to the bound steady state exhalation, $E_{\ell}(\infty)$. An important feature of the time-dependent diffusion theory is that this linear increase in radon concentration is dependent on the leakage factor, γ . (The very long reshaping times of thick samples, also giving linear growth rates of radon, are discussed below). The second question can be answered by looking at Equation 10 above. If the steady state exhalation, $E_{\ell}(\infty)$, is only slightly less than the free exhalation, E , or if the

free exhalation changes very slowly towards $E_{\lambda}(\infty)$, then the implication of the time-dependent diffusion theory may result in only minor corrections to 'old' measurements. If $E_{\lambda}(\infty) \simeq E$, it does not matter if we take a sample from the container soon after closure or later, as long as we correct the result for physical decay and leakage.

The concept of very long reshaping times is somewhat more intricate to explain. Consulting Equation 9 above, we see that the time-dependence of the exhalating system is given by the sum

$$\sum_i \exp -(y_i L/d)^2 \cdot \lambda_f t$$

Apparently there is little we can do about the y_i :s, at least for air-tight containers, the y_i :s attain values of about 2 or larger (cf Table 2 above). Only if the sample is thick compared with the diffusion length, L , will the first low-indexed terms in the sum above be 'slow' enough. For all thicknesses, d , larger than about $2L$, the difference between free exhalation rate, E , and the steady-state bound exhalation, $E_{\lambda}(\infty)$, will be the same, if other parameters are kept constant. In Table 4 are given the 10% depression times for thick samples. A radon-tight ($\gamma=1$) container is assumed. By a 10% depression time is meant the time it takes, upon closing the container, for the constrained exhalation rate, $E_{\lambda}(t)$, to deviate more than 10% from the free exhalation rate, E .

Thickness		Outer volume height	10% de- pression time
d cm	d/L	ℓ cm	hours
40	2	20	7.3
60	3	20	8.7
80	4	20	10.2
40	2	60	71
60	3	60	78
80	4	60	87

TABLE 4. The 10% depression time as a function of sample thickness, d, and outer volume height, ℓ. The diffusion length of the sample, is denoted by L. (Theory).

The conclusion to be drawn from the numbers given in Table 4 is obvious. Let us, for example aim at an accuracy of better than 10%. The time during which we have a constant exhalation, well approximating the free exhalation, is fairly long. Thus, sampling radon from the closed container, during say the first 24 hours, may for several 'thick' geometries be perfectly acceptable, when it is our intention to measure the free exhalation rate.

To summarize the issue of older measurements:

i. If the bound steady-state exhalation rate, $E_b(\infty)$, deviates very little from the free exhalation rate, ($pL \cdot \tanh(d/L)/\gamma\ell \ll 1$) the closed-can method yields the free exhalation rate as long as corrections for decay and leakage are applied.

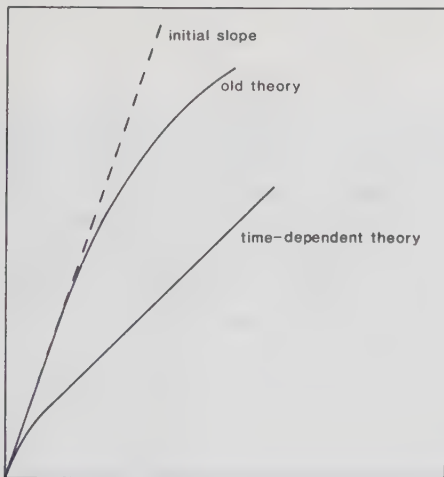
ii. For thick samples ($d > L$), the free exhalation rate is approximately obtained from the radon growth during the first few hours or several days, depending on the thickness and geometry, (cf Table 4).

iii. For thin samples ($d < 0.5L$), the closed-can method does not give the free exhalation, E , if the radon growth is measured after a few hours, due to short reshaping times. The mean exhalation rate obtained from a growth curve underestimates the free exhalation rate. The underestimation is less than a factor $(1 + pd/\gamma\ell)$, if leakage and decay corrections are performed. If only a decay correction is applied to a leaking accumulator, the free exhalation rate may be underestimated by more than the factor $(1 + pd/\ell)$.

What kind of experiments can be done to elucidate the special features of the

time-dependent diffusion transport of radon in porous samples? One possibility is to choose a sample thickness, d , and outer volume height, l , in such a way that the initial change in slope of the concentration as a function of time is pronounced. Choosing a thickness, d , less than the diffusion length, L , of the material will give a short reshaping time. A short reshaping time is an advantage, making the initial change in slope of the concentration distinct, and leaving plenty of time after the change, for checking the linearity in radon concentration growth. A linear increase corresponding to the bound steady state exhalation, if the time-dependent diffusion theory is valid. The initial course of the radon activity concentration in the outer volume, is schematically drawn for the old theory and for the time-dependent theory in Figure 3.

Radon concentration



Time

Figure 3. The old and new 'theories' of how the radon concentration increases upon closing the outer volume of an exhalation vessel. Schematic.

Characteristic of the old 'theory' is that the time-derivative decreases monotonously, due to what is believed to be an accelerating depression of exhalation. Typical for the time-dependent theory is that the initial change in slope is of short duration (for thin samples) and that the following activity concentration increase is linear in time, until decay losses of radon set in. Experimentally the curve shape predicted by the time-dependent diffusion theory is obtained, as exemplified in Figure 4.

Radon conc.
kBq m⁻³

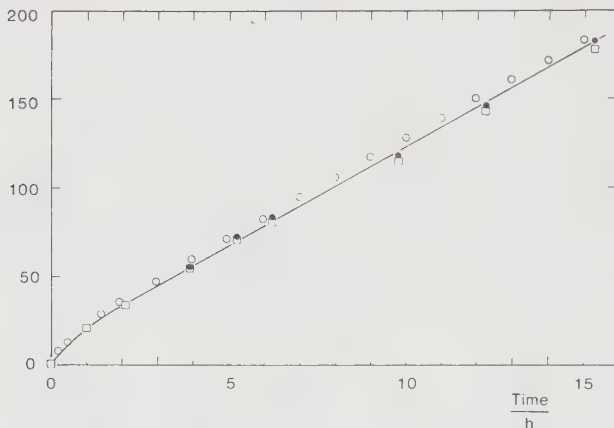


Figure 4. Radon concentration growth in the outer volume during the first fifteen hours after closure. The exhalating material is dry sand mixed with 11 % ground uranium ore by weight. The diffusion length, L , is 1.4 m, the sample thickness, d , is 26 cm and the outer volume height, ℓ , is 4.0 cm. (Porosity, $p = 0.47$, radium concentration, $C_{Ra} = 1180 \text{ Bq kg}^{-1}$, emanation fraction, $\epsilon = 0.33$, bulk density, $\rho = 1.71 \text{ kg dm}^{-3}$, relative leakage, $\gamma = 1$.)

- grab samples
- grab samples corrected for dilution effects.
- time-dependent diffusion theory,

Another way of testing the time-dependent diffusion theory is to measure the exhalation rate as a function of thickness, d , for thicknesses less than half the diffusion length, L . In the case of $d < 0.5 L$, the free exhalation rate is proportional to the thickness, d , and independent of diffusion length. The results of such a test are shown in Figure 4 in paper I. The deviation from linearity in thickness, d , of the experimentally obtained exhalation rate reveals that this rate cannot be a free exhalation rate.

Theoretical and experimental exhalation rates from a new series of tests are given in Figure 5. The exhalating material is sand mixed with 11 % ground uranium ore by weight. The porosity and diffusion length of this material, have been determined by independent methods.

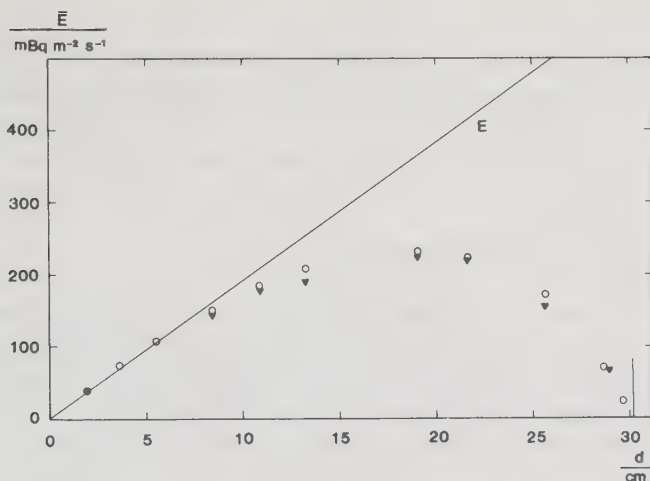


Figure 5. Theoretical and experimental exhalation rate as a function of sample thickness in an exhalation vessel of total inner height 30.5 cm. The sample parameters are identical to those in Figure 4.

— free exhalation, E .

○ time-dependent theory, mean exhalation rate during the first 10 hours.

▼ experiments, mean exhalation rate during the first 10 hours.

By filling the pores with ethanol, the porosity was deduced to be $47 \pm 2\%$ (± 1 s.d.). (The true figure may be somewhat higher if all the pores were not completely filled.) The porosity value is within the normal range for loosely packed sand-like materials. The diffusion length, L , is measured in the sand without uranium ore, to keep the self-production of radon low. The method used is similar to that described by Zapalac (Za83). The result from several experimental runs on samples of thicknesses between 0.1 and 0.2 m, is a diffusion length of about 1.4 m, which is a reasonable value, as the porosity is relatively high.

All samples included in Figure 5 and in Figure 4 (I) are thin in the sense that the thicknesses are less than $0.5L$. It is for samples in this category that the time-dependent diffusion theory has its greatest implications, due to the short reshaping times involved. In order to diminish the disturbing effects in the outer volume of air, the grab samples taken at sample thickness, $d = 26.5$ cm, have the volume of only 3.5 cm^3 .

There is a better agreement between theory and experiments in Figure 5, than in Figure 4 (I). There are several reasons for this. The dominant one is that the porosity used in Figure 4 (I), $p=0.40$, is too low. Furthermore the leakage during the series in Figure 4 (I) may have been higher than expected.

7.3 THE CONTINUOUS RADON MONITOR, (paper II).

A through-flow radon-in-air monitor is the subject of paper II. The theory of plate-out correction, in the simple form described in II, is straightforward. As a correction for the radon daughter plate-out is performed, the detector is designated a true radon monitor. Such a continuous radon monitor (CRM) is very useful when the radon concentration varies with time.

As seen in paper II, the time resolution of the CRM is limited. The 5-minute sampling interval yields correction coefficients, m_j , that are incorrect. Positive m_j values are physically impossible. The deficient m_j values are due to the finite wash-out time of radon from the CRM during calibration. A correction for the finite wash-out time is easy to apply, leading to a somewhat slower response of the detector. The 5-minute sampling interval is exploited infrequently by us, as the time resolution obtained using 15 or 30 minutes of pulse accumulation in many applications is sufficient. The microprocessor mentioned in paper II, has been exchanged. In the current CRM set-up, the correction is calculated by a Fortran program on a PDP 11/40 computer.

The continuous radon monitor is an excellent tool, when ever time-dependent radon levels are met. An example of this is given in Figure 6, where the monitor is utilized in an exhalation experiment, for the identification of grab-sample disturbances. The sand sample in Figure 6 is identical to the sample of the same thickness in Figure 5, but the exhalation run was carried out about four weeks later.

After taking the grab samples (Figure 6) the pressure drop in the outer volume was immediately compensated for by replacing air through a valve in the lid of the exhalating container. Dilution effects have not been corrected for in Figure 6. The small discrete samples of volume 3.5 cm^3 are subjected to larger uncertainties than the samples of normal volume taken directly into the ZnS flasks.

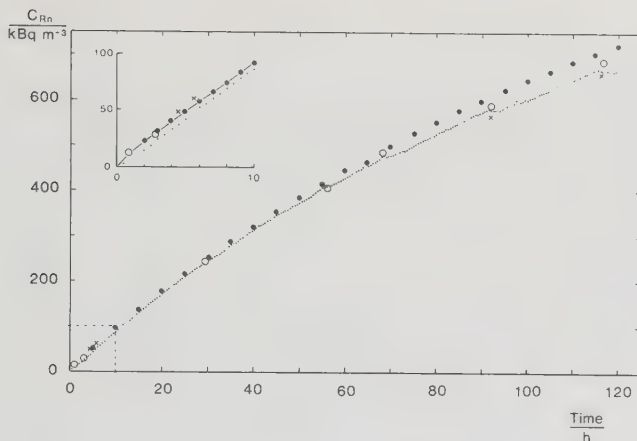


Figure 6. Theoretical and experimental radon concentration growth in an exhalation container. Sample thickness, $d = 21.5$ cm, see Figure 4 for other sample parameters. The CRM is a built-in type, used inside the exhalation container. The integration time period of the CRM is 15 minutes.

- Grab samples, volume 0.15 dm^3 .
- × Grab samples, volume 0.0035 dm^3 .
- Continuous radon monitor (CRM) results.
- Time-dependent diffusion theory.

The results in Figure 6 illustrate clearly that the grab sampling disturbs the smooth radon concentration growth, in spite of the steps taken to avoid pressure drops in the container. Further experiments are needed in order to better identify the sources of disturbances. One of these, the dilution of outer volume air, is significant as the outer volume is fairly small. For the 'thin' sample in Figure 6, the 'experimental' initial slope of the growth curve corresponds approximately to the bound exhalation rate, $E_{\ell}(\infty)$, as discussed in paper I and section 6.3 above.

In paper III, the CRM is connected to the radon room in order to check the constancy of the radon concentration. In this case the expected rate of change in the radon level is so slow, that the correction procedure described in paper II is of less importance. A sufficient accuracy is obtained from a simple linear relationship between the net count rate of the CRM and the radon concentration in the radon room.

7.4 THE EFFECTS OF AIR FILTRATION (paper III).

Embarking on the project described in paper III, the first goal was to find a simple and reproducible way of separating the unattached and the aerosol-attached short-lived radon decay products. It is shown in paper III, that the wire screen technique fulfills the need for simplicity and precision. But even though the wire screen method is convenient and simple to use, the collection- and alpha counting efficiencies must be carefully studied before starting practical separation measurements. It should be stressed that the calibration results obtained in paper III, cannot without further tests, be applied to radically different situations, for example high-humidity conditions.

A sample holder containing three wire screens in front of a membrane filter is used to measure the collection efficiencies for attached, a, and unattached, u, radon daughters. The experimental results are well supported by the theories of Cheng et al (Ch80, Ye82) and Thomas and Hinchliffe (Th72). The coefficients K_{ji} in Eq. 2 in paper III, can be plotted on a logarithmic scale, as a function of j . The shape of the K_{ji} curve obtained may be utilized to test the validity of the assumption behind Eq. 1 in paper III. In Figure 7, the relative K values are shown for RaA, RaB and RaC.

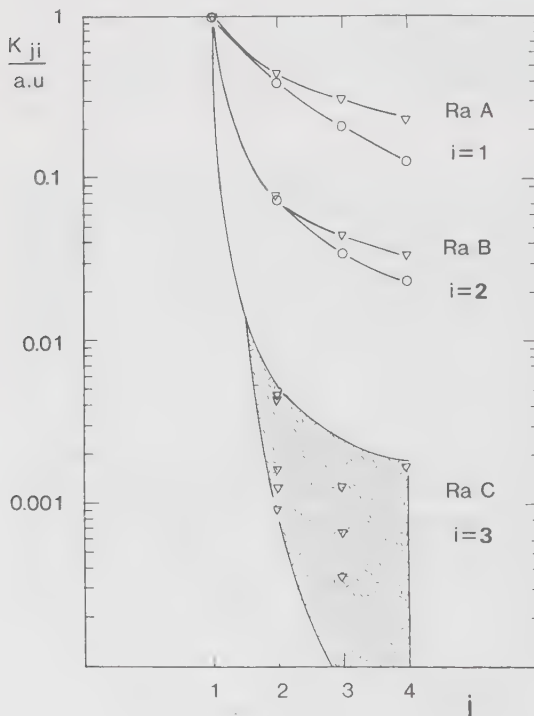


Figure 7. The relative wire screen coefficients, K_{ji} , (defined in paper III, Eq. 2). Typical curve forms during the condition when the unattached RaA and attached RaA are of the same magnitude. The wire screen is a 32 mm diameter screen of mesh 24 cm⁻¹.

∇ air sampling velocity interval 11-15 cm s⁻¹.

$\circ$ air sampling velocity interval 22-25 cm s⁻¹.

The results in Figure 7 are best interpreted by consulting Equation 2 in paper III. If the attached and unattached radon daughter concentrations are of equal magnitude, the quotient K_{4i}/K_{3i} is approximately equal to u , the collection efficiency of the wire screen for unattached activity. The RaA curves exemplify this in the Figure. If we assume that the collection efficiency, u , is close to 1, say 0.8, and the collection efficiency, a , for attached activity is small, say about 1 %, then the quotient K_{2i}/K_{1i} , is a good estimate of a , as long as the ratio unattached/attached activity is less than a . Such circumstances can prevail for RaC, during medium to high aerosol loads. The very large uncertainty (hatched region) in K_{ji} -values for RaC ($i=3$) and $j \geq 2$ is due to the very poor counting statistics. The slightly different aerosol conditions and sampling velocities behind the results in Figure 7, add further spread to the relative K_{ji} -values, for all three daughter nuclides.

One of the important aims of the investigation in paper III, is to evaluate the influence of air filtration on the effective dose equivalent rate. Two dose models are used for the calculations. One of them, the James-Birchall model, is deliberately chosen to represent models of high weighting factors for the unattached radon daughters. If the implementation of air filtration has favourable effects, according to the J-B model, it will also be favourable, from the dosimetric point of view, in models of lower weighting factors.

An important summary of the effect of air filtration on dose equivalent rate is given in Figure 19 in paper III. The large spread in the 'dose' reduction factor as a function of air cleaning rates in the filter is expected. Different virgin aerosols and different types of filters used are the cause. One important conclusion from Figure 19 (III) is that no increase in the effective dose equivalent rate is seen, not even in the James-Birchall model, weighting the unattached radon daughters by a factor of about 34. This is an important finding and it concerns for example people, who for reasons other than radiological ones, have to use air cleaners in their homes.

Another conclusion from Figure 19 (III), is that as a remedial tool, the effect of air filtration may be limited. In the practical range of filtration rate, say less than 5 h^{-1} , the reduction in effective dose equivalent rate is less than a factor of two or three (Fig. 19 III).

The Harley-Pasternak dose model (Ha72) used by Jonassen and McLaughlin (Jo82), uses a weighting factor of 26 for unattached ^{218}Po activity, compared with attached ^{218}Po , and dose contributions from unattached ^{214}Pb and ^{214}Bi - ^{214}Po are not included. The Harley-Pasternak dose model will therefore, in most cases,

give dose equivalent reductions somewhere in between the two models used in this investigation. In spite of the quite different experimental conditions, the air filtration results reported by Jonassen and McLaughlin show the same trend as those in Figure 19 (III). For intermediate aerosol loads (aerosol number concentration in the interval $3\text{--}5 \cdot 10^4 \text{ cm}^{-3}$) the dose reduction, obtained by Jonassen and McLaughlin using the Harley-Pasternak model, coincides essentially with the J-E curve in Figure 19 in paper III.

The concentration of attached and unattached radon daughters before and during air filtration at 5.2 h^{-1} by a high-efficiency filter, reported by Hinds et al, has been converted to effective dose equivalent rates by means of Equation 4 in paper III. The reduction in the effective dose equivalent is about 90%, relative to the air cleaner off conditions, in both the J-E and J-B models. The reason can be traced back to the fact that Hinds et al report a significant reduction in unattached ^{214}Pb and ^{214}Bi - ^{214}Po , after the onset of air filtration. This reduction is difficult to explain, considering the efficient aerosol removal by filtration, thereby decreasing the attachment rate considerably (cf Figure 1 and Table 3 above). The increased filtration- and plate-out losses of unattached ^{214}Pb and ^{214}Bi - ^{214}Po will, to a certain extent compensate for the failing loss through attachment. If an overcompensation is at hand, it will explain the reduced concentration of unattached ^{214}Pb and ^{214}Bi - ^{214}Po .

Actually, it is important to map the unattached RaB- and RaC concentrations, in spite of the often low absolute values. Both in the calculation of potential alpha energy (Eq. 4 above) and the effective dose equivalent (Eq. 35) the unattached RaB and RaC activities are assigned large weights. This can be illustrated by the most extreme case, comparing attached RaA and unattached RaB in the J-B model. For equal contributions to the effective dose equivalent the attached RaA activity must be a factor of 165 larger than the unattached RaB. The corresponding factor in the J-E model is 49.

Advanced aerosol measuring instruments are utilized in paper III. During the majority of the measuring series, both the aerosol size distribution and the condensation nuclei concentration, CN, were monitored. The size distributions cover the diameter interval $0.01\text{--}1 \text{ }\mu\text{m}$. The main purpose of introducing aerosol-measuring equipment, is to check the stability of the aerosol, in addition to the obvious desire to characterize the aerosol. Due to the detailed mapping of both the aerosol and the individual activity concentration, there is much more valuable information available in paper III than is discussed therein. The main objectives of paper III were to study the metrology of unattached activity and the subsequent use of the developed measuring technique to the air filtration

phenomenon. Therefore, discussions on subjects such as radon room models are deliberately excluded in paper III.

7.5 MEASUREMENT OF ^{222}Rn AND ^{210}Pb IN THE ARCTIC SUMMER AIR (paper IV).

A comprehensive description of the radiological part of the atmospheric chemistry programme of the Ymer-80 expedition is given in paper IV. Generally, equipment intended for indoor or laboratory measurements are utilized. Radon and the short-lived daughters are measured in situ, by more or less established methods. The inert radon is adsorbed onto cooled charcoal and the alpha activity concentration of the short-lived daughters are followed continuously by an alpha-in-air monitor, equipped with a membrane aerosol filter. These assessment techniques have been discussed in section 6 above. The air sampling on both the charcoal and the filter was performed in a 24 hour cycle, but the alpha count-rate of the filter was registered continuously by a data-logger. The '24 hour' filters for the short-lived daughters, were saved for later evaluation of the long-lived daughters ^{210}Pb and ^{210}Po .

Considering that the equipments are intended for laboratory use, and that personal relatively unfamiliar with the apparatuses, performed the sampling during the second leg, the reliability and continuity of the radon- and ^{210}Pb sampling were high. For radon only 5 days out of 78 were lost, and the corresponding figure for long-lived daughters is 2 out of 88 days. The on-line alpha monitor for short-lived radon daughters, was occasionally disturbed by sea salt and electronic noise. Because of such disturbances and human factors combined, about 30 % of the time was lost.

Due to the short preparation time and small budget the choice of instrumentation and parameters on board Ymer, deviate from the optimum. Though the experimental outcome of the expedition is more than satisfactory it is clear in looking back that improvements could have been made. The radon charcoal trap should have been larger, thereby avoiding leakage corrections. Also, the $\sim 50 \text{ m}^3$ air samples for long-lived daughters, are in the lower range of practical volumes. A larger sample is recommended, to improve the counting statistics when counting the activity from ^{210}Po .

The collection efficiency of the charcoal trap is shown in Figure 5 (IV). Since the printing of paper IV, the charcoal trap has been studied with regard to the multi-compartment theory described in section 6.1. The evaluation of these dynamic studies of the charcoal-adsorbing characteristics is now underway. One

experimental point with parameter values similar to those used during the Ymer-80 expedition has been determined. At a trap temperature of -31°C and an air flow rate of 1.3 l min^{-1} , the breakthrough volume is about 700 litres and the collection efficiency is 79% after a collection time of 22 hours (corresponding to 1.7 m^3 in this case). This collection efficiency is slightly below the efficiency curve (85%) in Figure 5 (IV), obtained at temperatures between -35 and -40°C .

A huge amount of data describing the arctic atmosphere was gathered during the expedition. Only a small part of this valuable information is described in paper IV, with emphasis on the radiological part of the air chemistry programme. All radon concentration values obtained during the expedition are shown in Figure 6 (IV), and examples of air trajectories are given in Figure 7 (IV). Apparently there is a strong negative correlation between air-mass age over the sea and radon concentration. The sea-level trajectory is the most important trajectory determining the radon concentration. All high radon levels, are measured in 'young' air (cf e.g. trajectories 6 and 9 in Figure 7 in IV). For all locations north of 75°N the mean radon concentration during the expedition was $75 \pm 21\text{ mBq m}^{-3}$ ($\pm 1\text{ s.d.}$). This value is low compared with radon concentrations reported from the Alaska region (Lo62, UN82) and the Norwegian sea (UN82), indicating a very low influence from continental air masses. The low radon concentrations found, support the idea of Rahn (Ra78) that the Arctic region is decoupled from the midlatitudinal regions during the summer. The ^{210}Pb concentrations measured are within the range found on Franz Josefland (Ra76), but the grand mean value of the Ymer-expedition, $75\text{ }\mu\text{Bq m}^{-3}$, is much larger than the mean value, $13\text{ }\mu\text{Bq m}^{-3}$, reported by the Russians. The ^{210}Pb concentration is strongly dependent on precipitation intensity and frequency. The large deviation of the mean ^{210}Pb concentrations quoted, obtained during different time periods, may therefore be due to differences in meteorological factors.

A part of paper IV is devoted to an analysis of potential disturbing factors. It is shown that both the radon and the ^{210}Pb measurements are insensitive to contamination from the ship itself and from local settlements. The sea-salt causes no radioactivity contamination either, but can prohibit the proper functioning of the surface-barrier detector. One local radon source that may be of importance is the sea. Crude calculations based on the gas-exchange data reported by Roether and Kromer (Ro78), cannot exclude the sea as a significant radon emitter during the low concentration period in July, showing a mean value of about 20 mBq m^{-3} . The insensitivity of the radiological measurements to the ship's emission is a great advantage, making elaborate contamination controlling systems unnecessary.

Activity ratios between the different long-lived radon daughters, and between long-lived radon daughters and radon itself, may be used to calculate the mean residence time of the aerosol in the atmosphere, above the continents, as discussed in section 3.2.3 above. Steady-state models are used, and due to the large variation in radon source strength, the mean residence times in the atmosphere over land, are uncertain by a factor of 2-3 (Ju63). The ratio of ^{210}Po to ^{210}Pb is not recommended for use in residence time calculations, as sources other than atmospheric radon exist for ^{210}Po (Ma74). The activity ratio $^{210}\text{Bi}/^{210}\text{Pb}$ is proposed as the ratio best suited to such calculations (Ma74). In paper IV it is shown (cf Figure 12 in IV) from basic decay- and elimination principles, that no steady state is achieved in the air over the sea, unless the mean residence time of the aerosol is much less than the mean residence time, 5.5 days, of radon. In all cases when the true residence time is larger than about 1 day, the use of simple steady-state formulae presupposing a long-lived mother (cf Eq. 17 in section 3.2.3 above) overestimates the true residence time. This also applies to the recommended ratio $^{210}\text{Bi}/^{210}\text{Pb}$, if measurements are taken, as assumed above, in the air over the oceans.

Episodes of long-range transport to the Arctic region, and episodes of local contamination are also discussed in paper IV. In air masses originating from the continents, there is a close correlation between short-lived radon daughters (RnD) and the light-scattering coefficient, σ_{sp} , a measure of the aerosol mass in the size range 0.2-1 μm (Figure 12 in IV). The emissions of local settlements have a low radioactivity content. Accordingly, the radioactivity concentrations will not correlate with either the condensation nuclei concentration, CN, nor the light-scattering coefficient, σ_{sp} . A comparison between the radon- and radon daughter concentrations and the stable constituents such as sulphur, chlorine etc., is difficult due to the largely differing sampling time intervals. In spite of these difficulties, it can be said that the Ymer results support the hypothesis that larger industrialized land masses are sources of both radon and stable pollutions, like sulphur. Non-industrialized areas emit radon, while sulphur emission is nearly absent. These conclusions are exemplified by the long-range transport incidences showing high values of both radon and sulphur. Greenland, on the other hand, is a source of radon during the summer, but is an insignificant source of sulphur. All the comparisons with stable pollutants and values of, for example, condensation nuclei concentration, CN, and light-scattering coefficients σ_{sp} , have been made possible thanks to the paper published by Lannefors et al (La83) and to personal communications with these authors and Dr Lennart Granath (Gr84) at Stockholm University.

8. SUMMARY

Several aspects of radon (^{222}Rn) and its decay products are included in this investigation. Radon and radon-daughter measurement methodologies are analysed from both theoretical and experimental points of view. It is shown that the results obtained from exhalation measurement can be explained by diffusion being the only transport mechanism. Deficiencies in the established accumulation method of radon exhalation measurement are shown by the use of the time-dependent diffusion theory. The term 'back-diffusion', hitherto vaguely defined, is clarified and shown to be applicable only to steady-state conditions.

Enclosing a thin (with dimensions less than half the diffusion length) porous sample in an accumulating vessel leads to a rapid change from a free to a bound (constrained) exhalation rate. The nearly linear radon-concentration growth in the outer volume of the vessel in such a geometry is shown to underestimate the free exhalation rate by a factor up to $(1 + \alpha^{-1})$ in radon-tight vessels (α is the outer-to-inner volume ratio.)

The wire-screen technique combined with surface barrier detectors is a convenient and reproducible method for separation of aerosol-attached and unattached radon-daughter activities. The effect of air-filtration on the radon-daughter levels are in this investigation expressed both in activity and in dosimetric (effective dose equivalent) terms. Air-filtration of indoor air has no deteriorious effects, even when evaluated by dose models, weighting the unattached fraction of radon-daughter activity by the largest factors. That the implementation of air-filtration is worthwhile as a remedial counter-measure in 'radon' houses can not be generally asserted, as the initial radiological, economical and social factors are too diverse. In this investigation, the effective dose equivalent rate has been reduced by a factor of 1.3 to 2.5, depending on initial aerosol conditions and the dose model used, at a filtration rate of 5 h^{-1} . Fine filters, ordinary and of type electret, of surface densities in the range $0.6\text{--}3.2 \text{ g cm}^{-2}$, have been used.

The grand averages of ^{222}Rn obtained during the Ymer-80 expedition are $75 \pm 21 \text{ mBq m}^{-3}$ ($\pm 1 \text{ s.d.}$), and the average ^{210}Pb concentration is lower by a factor of 1000. It is shown theoretically that long-lived daughter activity ratios are unsuitable as an estimator of the aerosol residence time in ocean air. A good qualitative agreement between radon-levels and the time since the air left larger land areas was found. The measurement of ^{222}Rn and its long-lived daugh-

ters are insensitive to contamination from the ship's exhaust-emission and emission from local settlements. During Arctic background conditions, significant contribution from sea-emanating radon cannot be excluded.

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EXHALATION OF ^{222}Rn FROM
POROUS MATERIALS.

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ABSTRACT

The theory of time-dependent radon diffusion transport has several implications on exhalation measurement techniques. It is shown that a one-dimensional theory is applicable to samples in containers if the wall of the container is non-absorbant. A reshaping of the internal radon concentration within the sample is responsible for the transfer from free to bound exhalation upon closing the container. The reshaping time is defined as the time taken to reach steady-state exhalation. Examples are given where the reshaping times are so short that the steady-state bound exhalation completely dominates the radon concentration growth in the outer volume. In such cases the mean exhalation rate during the first hours may be a gross underestimation of the free exhalation rate. As the steady-state exhalation rate depends on the air leakage rate of the container, the leakage term must be known. It is stressed that the concept of 'back diffusion' is applicable only in the description of how the bound steady-state exhalation diminishes with decreasing outer air volume in an enclosure.

INTRODUCTION.

Problems concerning the radon-222 transport processes are met both in outdoor and indoor environments. In houses the source of radon-222 can be the building material itself, materials surrounding the house or supplied to the house. The radon-222 decay products are a major source of natural background radiation in several countries⁽¹⁾.

The main object of this paper is to present a discussion on the diffusion theory applied to radon exhalation and its implications in experimental measuring techniques.

THEORY.

The release of radon to the surrounding air from a porous body (exhalation) is a two-step process. First the emanation of radon to the pore system and secondly the transport through the pores (or cracks) to the surface of the body. The driving forces in the transport can be pressure or concentration gradients.

Capillary transport of radon in partly water-filled pores may also be of significance. Only the theory of diffusion will be described here.

If two dimensions of a sample are much larger than the diffusion length, a one-dimensional analysis is applicable. In such a semi-infinite sample there is no net transport in the x- or y- directions. A concentration gradient exists only in the z-direction. Imagine a subsample in form of a cylinder (Fig. 1). As there is no net flow through the cylinder wall the diffusion in the subsample within the cylinder is also one-dimensional. This means that samples within cylinders (or any other straight-walled duct) fulfil the requirements of the one-dimensional treatment as long as there is no erroneous radon flow along the inner surface of the wall; or at least that such a flow ('wall effect') is negligible compared with the bulk diffusional flow. By similar reasoning it is clear that the demand that the sample be semi-infinite can also be relaxed in the case of a sample enclosed in a canister in the absence of 'wall effects' (cf Fig. 1b).

For a semi-infinite porous block of thickness $2d$, covered by lids on both sides, or a subsample of thickness d in a canister, the differential equation for diffusion is

$$\text{Eq. 1} \quad \frac{\partial C(z,t)}{\partial t} = \lambda_f C_m - \lambda_f C(z,t) + D \frac{\partial^2 C(z,t)}{\partial z^2}$$

$C(z,t)$ = concentration of radon in the pores at time t (Bq/m^3)

C_m = maximum value of C in an infinite sample.

D = diffusion coefficient for radon (m^2/s)

p = porosity, the fraction of the sample volume occupied by air.

λ_f = decay constant of radon (s^{-1})

The diffusion coefficient in equation 1 is connected to the molecular diffusion coefficient D_m of radon in air by the relation $D = hfD_m/p$, where

h = tortuous factor

f = fraction of a cross sectional area in the x-y plane occupied by open pores.

(Culot et al⁽²⁾ have discussed different ways of defining effective diffusion coefficients). The radium-226 content, C_{Ra} , (Bq/kg) of the sample is related to C_m by the relation $pC_m = C_{Ra} p\epsilon$, where

ϵ = emanation factor, the fraction of the radon produced in the material that reaches the pore space.

ρ = bulk density of the sample (kg/m^3)

The boundary conditions are:

$$\text{Eq. 2} \quad \frac{\partial C(z,t)}{\partial z} = 0 \quad \text{when } z=0$$

$$\text{Eq. 3} \quad -Dp \frac{\partial C(z,t)}{\partial z} - (\lambda_f + \lambda_v) \cdot l \cdot C(z,t) = l \frac{\partial C(z,t)}{\partial t} \quad z=d$$

where l is the air height above the sample and λ_v is the ventilation rate constant of the enclosure. Equation 3 is the mathematical expression for the balance between the exhalation (first term) and the loss of activity (second term) due to decay or ventilation. If there is imbalance, the activity concentration at the interface ($z=d$) will change with time. Equation 3 is not based on the equality between p and f , but requires an instant air mixing in the outer volume. When $\partial C/\partial t=0$ we have the steady-state solution:

$$\text{Eq. 4} \quad C_1(z,\infty) = C_m - C_m \frac{\cosh(z/L)}{\cosh(d/L) + \frac{pL}{\gamma l} \sinh(d/L)} \quad \text{where}$$

C is indexed by l denoting that the air height above the sample is equal to l . $\gamma = (\lambda_f + \lambda_v)/\lambda_f$ is the relative leakage. The diffusion length L is equal to $(D/\lambda_f)^{1/2}$. The bound exhalation at steady-state ($t \rightarrow \infty$) is given by Fick's law

$$\text{Eq. 5} \quad E_1(\infty) = -Dp \left. \frac{\partial C(z,t=\infty)}{\partial z} \right|_{z=d} = \frac{C_m p \lambda_f L}{\coth(d/L) + \frac{pL}{\gamma l}}$$

The ratio between the free ($l \rightarrow \infty$), E , and bound steady-state exhalation E_1 is

$$\text{Eq. 6} \quad \frac{E}{E_1} = 1 + \frac{pL \tanh(d/L)}{\gamma l}$$

Suppose we have a block in equilibrium exhaling into free space and that we enclose the sample at time $t=0$. The initial conditions for the concentration gradient within the block are given by Equation 4 as $l \rightarrow \infty$. The free exhalation E at time zero will gradually change towards the bound exhalation E_1 . To see how fast this change in exhalation will take place we have to look at the time dependent solution of Equation 1. The solution is (3)

$$\text{Eq. 7} \quad C(z,t) = C_m \left\{ 1 - \frac{\cosh(z/L)}{\cosh(d/L) + \frac{pL}{\gamma l} \sinh(d/L)} - \frac{\tanh(d/L) \{1 + \alpha \frac{d}{L} \coth(d/L)\} \exp(-\lambda_f t)}{\frac{d}{L} (1 + \alpha) \{1 + \gamma \alpha \frac{d}{L} \coth(d/L)\}} - G(z,t) \right\}$$

where $\alpha = l/pd$ is the ratio between outer and inner volume. It should be noted that the first two terms are equal to $C_1(z, \infty)$ and that the third term is independent of z , i.e. makes no contribution to the exhalation. The fourth term in Equation 7 describes how the shape of the concentration curve within the block transforms to congruency with the concentration gradient corresponding to bound exhalation. The exact expression for the function $G(z, t)$ when $\gamma = 1$ is

$$\text{Eq. 8} \quad G(z, t) = \frac{2 \tanh(d/L) \cdot \exp(-\lambda_f \cdot t)}{d/L} \cdot$$

$$\sum_{i=1}^{\infty} \frac{d^2 \cdot \cos(y_i \cdot z/d) \cdot \exp\{-(y_i \cdot L/d)^2 \cdot \lambda_f \cdot t\}}{(1 + \alpha + \alpha^2 y_i^2)(d^2 + y_i^2 L^2) \cos y_i}$$

where y_i solves the transcendental equation $-\alpha y_i = \text{tg } y_i$. (The roots y_i increase monotonically with a difference approximately equal to Π , starting with $\Pi/2 < y_1 < \Pi$). The course described by Equation 7 is illustrated in Figure 2. In this case the fast disappearance of the free exhalation rate should be noted. Experimentally the small bend at the beginning of the concentration curve will pass unnoticed.

DEDUCING EXHALATION RATE FROM RADON GROWTH.

Radon exhalation measurement are mainly based on the principle of enclosing or covering the sample to be measured. Different techniques are then utilized for collecting the radon from the enclosure. The aim of this section is not to discuss the merits of different exhalation measurement methods, but, in more general terms, to illustrate what is actually measured when sampling radon from an enclosed specimen. The discussion is limited to one-dimensional geometries (cf Fig. 1) and the sampling and radon detecting system are assumed to be ideal. From equation 6 it is clear that the free exhalation is approximately equal to the bound exhalation E_1 if $\{pL \cdot \tanh(d/L)\} / \gamma L \ll 1$, and for thin samples ($d < 0.5L$) in tight canisters it is simply $\alpha^{-1} \ll 1$. If a controlled leakage is applied to the canister the requirement for a large outer to inner volume ratio can be relaxed. If the inequality above is fulfilled, the free exhalation rate is easily determined, either from the initial slope of the radon concentration versus time curve or from a steady-state determination. In both cases the leakage coefficient λ_v must be known.

It is important to note that in several experimental situations the bound steady-state exhalation has a major influence on the slope estimated from the radon concentration growth in the enclosure. Thus the hypothesis that 'as long as the radon increase appears linear in time, it corresponds to the free exhalation' is

wrong. In the case of the air-tight canister illustrated in Figure 2, the free exhalation rate corresponds to a radon concentration 36 % higher, at $t=500$ minutes, than the value actually attained. If, for instance, a single air sample is taken after 500 minutes, the mean exhalation rate assessed, seriously underestimates the free exhalation rate. The phenomenon is also illustrated in Figure 3, but for a shorter diffusion length. In the left-hand part the time-dependent exhalation per unit mass, as derived from Equation 7, is displayed ($\gamma=1$). The right-hand part of the figure shows the corresponding radon concentration growth. For the two samples, with thicknesses of 10 and 20 cm, the theoretical radon concentration increase for free exhalation without decay, free exhalation corrected for decay and for the time-dependent bound exhalation are shown.

In the literature the depression of the exhalation rate, experienced when enclosing a specimen in a canister, is often attributed to 'back diffusion'. It is obvious that most authours are of the opinion that back diffusion is progressive with radon concentration in the outer volume, and as long as the concentration in this outer volume is insignificant compared with the 'pore concentration', the back diffusion is negligible. By consulting the example in Figure 2 it is clear that 'back diffusion' in the sense of depressing the exhalation rate, takes place immediately after closing. Once the bound steady state exhalation is reached, (in Fig. 2 after about 150 minutes) any further increase in the outer radon concentration has no effect on the exhalation rate. When designating the cause of the exhalation decrease as a function of time, it is evident that the term 'back diffusion' is misleading and should not be used. Instead the word 'reshaping' is proposed, as it is the reshaping of the internal concentration gradient that causes the reduction. The time taken for the system to change from free to bound steady-state exhalation is consequently called the reshaping time. To be precise when giving reshaping times as numerals, it is necessary to state which percentage (e.g. 95%) of ΔE ($\Delta E = E - E_1(\infty)$) one is referring to. Reshaping times on the 95% level are given in Table 1.

EXHALATION AND SAMPLE THICKNESS.

If specimens of different thicknesses are enclosed in identical containers, Equation 5 implies that the bound steady-state exhalation rate, $E_1(\infty)$, is ambiguous as a function of thickness. Samples of two different heights may result in the same $E_1(\infty)$. In the case exemplified in Figure 4 the maximum bound exhalation is reached at a thickness of about 14 cm. Above 14 cm the high radon concentration in the container lowers the absolute value of the bound steady-state exhalation. If instead the sample thickness is kept constant and the height of the outer volume l is decreased towards zero, a monotonically diminishing steady-state exhalation rate will result. In our opinion it is pertinent to reserve the term 'back diffusion' for the phenomenon just described. It

should be observed that in this definition of back diffusion, there is no time dependence, as the term refers to steady-state exhalation. The exhalation is zero at the limit $l=0$, which we have exploited above during the discussion of one-dimensional geometries (cf Fig. 1).

EXHALATION EXPERIMENTS.

At the time of writing no experiments have been designed specifically to test the theoretically derived results above. An old series of measurements however has been reproduced in Figure 4 for a comparison with theory. A mixture of sand and 3 % ground uranium ore was stored in the laboratory for acclimatization and then poured into a canister of diameter 27 cm and height 22.7 cm. Before each exhalation run the sample was left in the open canister for at least 24 hours to reach equilibrium. Upon closing the canister the radon growth was followed by taking 4-6 discrete air samples with pre-evacuated ZnS chambers within the first 12 hours. An apparently linear radon concentration increase was obtained, and the corresponding mean exhalation rate corrected for decay and leakage ($\bar{E}_{\text{exp}}$) was calculated. Assuming a diffusion length of about 1 metre or larger, the decrease of $\bar{E}_{\text{exp}}$ per unit mass is not expected if free exhalation is measured, but is predicted by the time-dependent diffusion theory (curve $\bar{E}_1(10h)$) in Fig. 4). At the moment we have no clear explanation for why the experimental values, $\bar{E}_{\text{exp}}$, are significantly lower than those given by theory. Several possible sources of error exist. The porosity is not accurately known, and the leakage factor is not determined for the specific canister used. Furthermore, the theoretical demands on one-dimensional exhalation and on immediate air mixing in the outer volume may be violated, as the wall effect has not been investigated and as the air in the canister has not been stirred. It is obvious that carefully designed exhalation experiments need to be done to guarantee the validity of the time-dependent diffusion transport theory in porous materials.

CONCLUSIONS

To understand and clarify exhalation phenomena it is necessary to solve the time-dependent differential diffusion equation. From the solution it is evident that a reshaping of the internal radon concentration gradient is responsible for the transfer from free to bound exhalation. In many cases the reshaping times are so short that it is more or less impossible to experimentally deduce the free exhalation rate from the initial growth of the radon within an enclosure. Staying with the old definition of 'back diffusion' as the cause of a reduction in exhalation caused by a high radon concentration in the outer volume, it is concluded that the concept is irrelevant when a single specimen is enclosed in a given geometry. In such a case the term 'reshaping' is proposed instead. Back

diffusion is relevant however as a cause of the lowering of the steady-state exhalation rate from a specimen when the outer volume decreases. Further experimental validation of the theoretical model described in this paper is needed.

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FIGURE CAPTIONS.

1. a) A semi-infinite slab of thickness $2d$ exhaling into free space.
b) A subsample of thickness d in an enclosure of total height $l+d$.
2. Results of theoretical calculations with parameter values:
 $C_m = 75 \text{ kBq/m}^3$ $d = 15 \text{ cm}$ $l = 15 \text{ cm}$
 $L = 100 \text{ cm}$ $\gamma = 1$
 The mass of the sample is 13 kg.
 a) The internal radon concentration in the sample.
 b) The free exhalation E and the bound time-dependent exhalation E_1 .
 c) The radon concentration C in the outer volume (the lower curve is corrected for physical decay) corresponding to E and the radon concentration C_1 corresponding to E_1 .
3. Similar to Figure 2b and 2c, but for diffusion length $L = 20 \text{ cm}$, $l+d = 22.7 \text{ cm}$ and three different thicknesses. The specific exhalation and concentration are shown. The mass density is 1.5 kg/dm^3 . For thickness $d = 5 \text{ cm}$ only the radon concentration corresponding to time-dependent exhalation is plotted.
4. Theoretical and experimental exhalation as a function of sample thickness in a canister of total height $l+d = 22.7 \text{ cm}$.
 The sample is a mixture of sand and 3% ground uranium ore.
 $C_m = 75 \text{ kBq/m}^3$ and $\gamma = 1$.
 $E =$ free exhalation
 $E_1(\infty) =$ bound steady-state exhalation
 $\bar{E}_1(10h) =$ mean exhalation during $t = 0-10 \text{ h}$
 $\bar{E}_{\text{exp}} =$ experimental exhalation as deduced from the radon concentration increase during $t = 0-12 \text{ h}$.

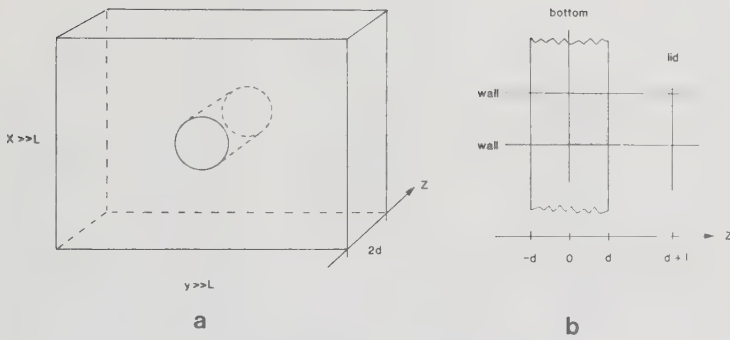
TABLE CAPTION.

- 1 The 95% reshaping time (see text) for different combinations of sample thickness d , canister height $l+d$ and diffusion length L . An air-tight enclosure ($\gamma=1$) is assumed.

TABLE 1.

Sample Thickness	Outer volume height	Canister height	Diffus. length	Free to bound steady state exhalation	Reshaping time
d	l	l+d	L	$E/E_1(\infty)$	t(95%)
cm	cm	cm	cm		minutes
2	48	50	100	1.02	5
10	40	50	100	1.13	80
40	10	50	100	2.90	570
2	48	50	20	1.02	70
10	40	50	20	1.12	1840
40	10	50	20	1.96	6260
2	13	15	100	1.08	4
10	5	15	100	2.00	50
2	13	15	20	1.08	90
10	5	15	20	1.92	1180

Figure 1



- a) A semi-infinite slab of thickness $2d$ exhalating into free space
 b) A subsample of thickness d in an enclosure of total height $l+d$.

Figure 2

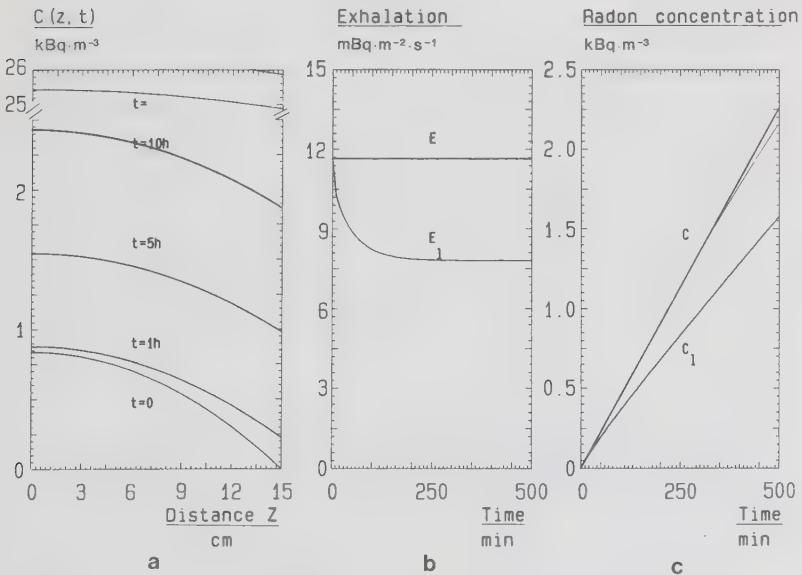


Figure 3

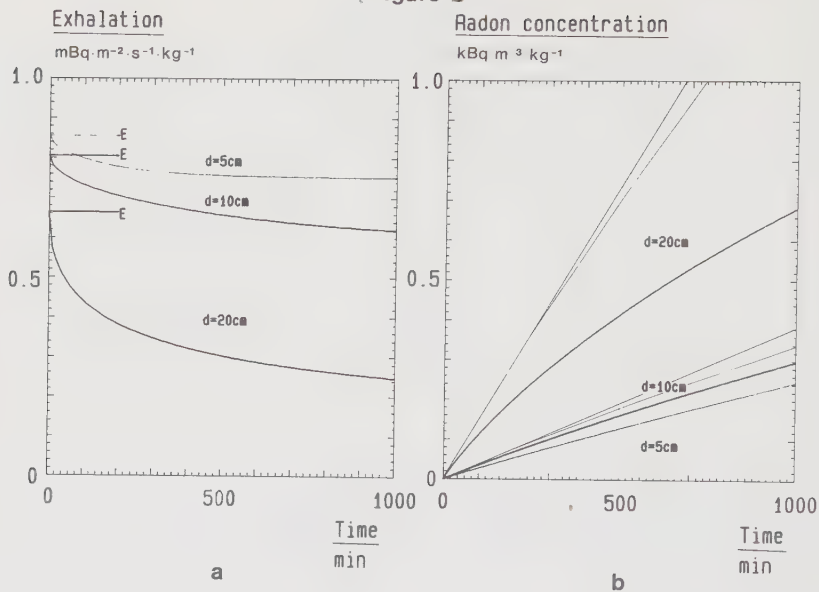
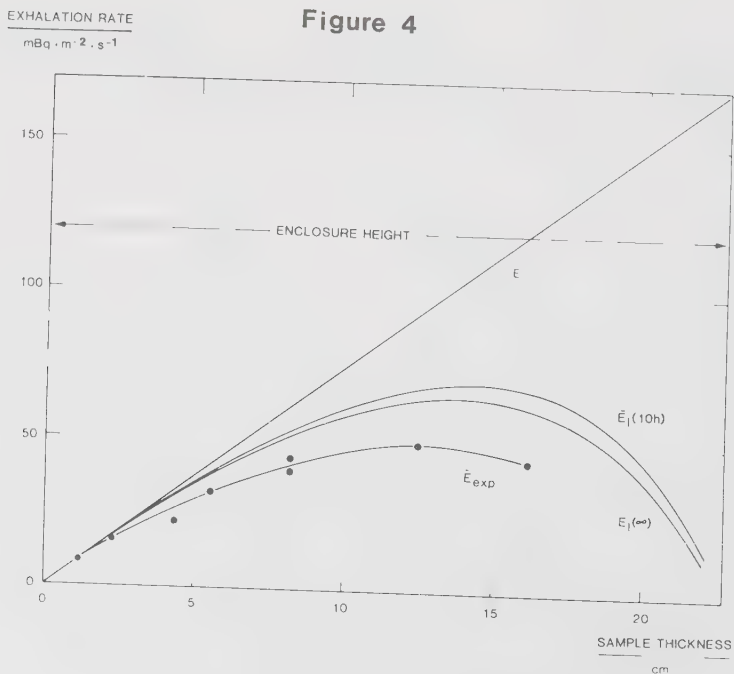


Figure 4



A CONTINUOUS TRUE RADON MONITOR.

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ABSTRACT

A continuous radon monitoring system has been constructed where the memory effect is eliminated by multiplying the number of pulses in each time interval with a correction function. The essential components of the system are the flow-through detector, a zinc sulphide cell attached to a PM-tube, and a microprocessor.

The microprocessor stores the number of pulses in consecutive time intervals. The true radon concentration in each time interval is calculated by the microprocessor and printed out on a printer. Four different time intervals are used, from 5 to 90 minutes, giving four different 'time constants'.

The time resolution of the system has been measured by exposing the detector to radon pulses of different lengths. The response is in good agreement with the true radon concentration provided the radon pulse duration is long compared to the time constant used. A sensitivity down to 20 Bq/m³ radon is achievable with a low background ZnS-cell.

1. INTRODUCTION

Several continuous radon level monitors have been constructed during the years. We will confine the discussion to flow through radon detector cells and exclude measuring devices based on the principle of deducing the radon concentration from a measurement of its short-lived progenies on filters. Ionization chambers or scintillation cells have been used as detectors. In these cases, the whole flow cell constitutes the detector. The scintillation cell system is more suitable for field measurement because of the smaller size of the detector. Another possibility is to place a continuously registering detector inside a non-sensitive flow chamber. In all applications, the radon laden-air is prefiltered to remove radon daughters. Due to the finite residence time of air in the chamber, radon daughters are formed in the chamber. These daughters plate out on surfaces (in some applications, deliberately, by means of an electric

field or are swept out by the air stream. The plated-out decay products give a delayed response, characteristic for all the types of detectors described above.

2. THEORY OF RESPONSE.

If the flow through chamber is exposed to a constant level of radon and then given radon-free air, the wash-out curve of the detector is obtained. This wash-out includes both effects of physical build-up and decay of radon daughters in the chamber and the wash out of radon and daughters by the air stream. Under the assumption that the radon concentration C is constant during any time interval, the signal S received in time interval n can be written:

$$(1) S_n = (\epsilon_r + \epsilon_d) C_n + \epsilon_d k_1 C_{n-1} + \epsilon_d k_2 C_{n-2} + \dots$$

The ϵ_r and ϵ_d are the counting efficiencies (counts per Bq/m³) of radon and radon daughter products respectively. k is the elimination coefficient of radon daughters in time interval i . The wash out and decay of the radon daughters are complicated processes involving components of build-up from radon, plate-out on surfaces, recoil from surfaces etc. Equation 1 is an oversimplification, as it puts all these components into a single elimination coefficient, k . In Equation 1 it is also assumed that the radon gas is washed out instantaneously. This assumption imposes the restriction that the time interval used must be long compared to the residence time of radon in the detector.

In a mean level-ionization chamber, the signal is the charge collected in a time interval and in a pulse-type ionization chamber or a scintillation cell, it is the number of pulses in an interval. A digital signal is easier to treat mathematically and we assume in the following analysis that the signal is digitalized. It is not necessary to know all of the physical details to deduce the true radon concentration, C , from the number of pulses, S . One must, however, know the number of pulses in all time intervals preceding and influencing interval n . Roughly, all intervals up to three hours before interval n must be considered. If we in Equation 1 put the efficiencies and the elimination factors together we can write it in the simple form given by Thomas (1):

$$(2) S_n = \epsilon(C_n + l_1 C_{n-1} + l_2 C_{n-2} + \dots + l_i C_{n-i} + \dots)$$

where $\epsilon = \epsilon_r + \epsilon_d$ and $l_i = \epsilon_d k_i / \epsilon$

From a set of Equations (2) giving the signals in time intervals preceding interval n , we can successively eliminate C_{n-1} , C_{n-2} etc. to get:

$$(3) C_n = (1/\varepsilon) (S_n + m_1 S_{n-1} + m_2 S_{n-2} + \dots + m_i S_{n-i} + \dots)$$

The elimination algorithm is given by the sum:

$$(4) m_j = - \sum_{i=1}^j l_i m_{j-i}$$

and the coefficients, m , can be calculated iteratively, starting from $j=1$ and $m_0=1$. The coefficients, m , must be determined for each flow rate and each time interval used. This disadvantage reflects the empirical 'not-knowing-the-physical-details' approach in Equation 1.

3. CALIBRATION

By exposing the detector cell to known levels of radon, the coefficients, l , and the efficiency, ε , in Equation 2 can be deduced. The memory coefficients, m_i , in Equation 3 is then calculated by Equation 4. If the radon concentration is kept constant for at least three hours, and the detector is then fed with radon-free air at the start of time interval $n+1$ and onwards, we have from Equation 2 ($l_0 = 1$):

$$(5) S_{n+i} - S_{n+i+1} = \varepsilon l_i C_n \quad i = 0, 1, 2, \dots$$

By taking grab samples in pre-evacuated and calibrated ZnS-cells during the constant period the radon concentration C can be measured. Then the efficiency and the coefficients l can be calculated from Equation 5. If the radon instead is given as a pulse during the time interval, n , we have:

$$(6) C_i = \begin{cases} C_n & i=n \\ 0 & i \neq n \end{cases}$$

l_i and ε are calculated according to $S_{n+i} = \varepsilon l_i C_n$, where S_j is the signal (number of pulses) in time interval j . In the above calculations we have assumed that the signals are corrected for detector background.

The countrate decay following a constant level of radon is exemplified in Figure 1 for the time intervals 5 and 30 minutes. From these experiments, the coefficients m have been calculated for four different time intervals, 5, 15, 30 and 90 minutes. The background signal of the ZnS-cell used in Figure 1 is unusually high (7 cpm). The main reason for this is the contamination of the detector by long-lived alpha activity. The cell had, prior to the experiment shown in Figure 1, been used for a long time in atmospheres with high radon concentration.

4. CONSTRUCTION

A continuous radon monitoring system has been designed to measure the true radon concentration in air. The essential components are the flow-through detector, a zinksulphide cell at-

tached to a PM-tube, and a microprocessor. The inlet air is cleaned from dust and radon daughters by a membrane filter. Conventional pulse-amplifying electronics and a pump complete the system. The number of pulses in consecutive 5 minute-intervals is stored by the microprocessor. The radon concentration, C , in a time interval is calculated from Equation 3 by the processor and printed out together with the corresponding pulse number. The sensitivity of the system is 1.6 cps per kBq/m^3 , and the background about 0.02 cps. At low levels of radon, the number of net pulses is too small, 1 pCi/l (37 Bq/m^3) gives about 20 pulses in 5 minutes, to give a reasonable accuracy in the calculated true radon values, C . The time intervals 15, 30 and 90 minutes for pulse accumulation are therefore also treated by the microprocessor, and the corresponding true radon values calculated. A longer time interval gives higher statistical accuracy in the individual C -value at the expense of a poorer time resolution.

Count rate
 a.u.

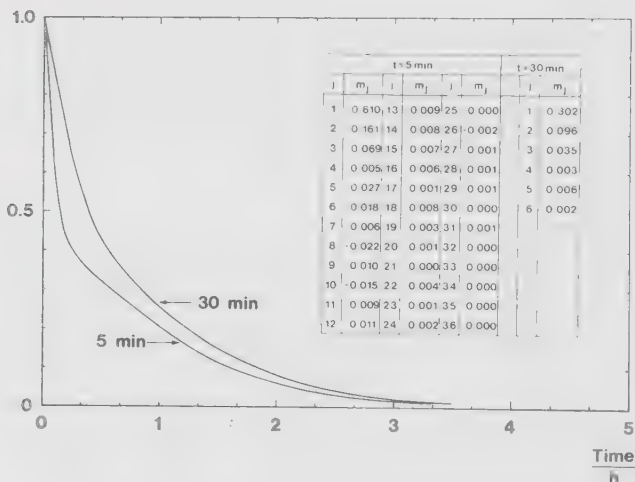


Figure 1. The wash-out curve of the detector after a constant level of radon. The air flow equals $1.8 \text{ dm}^3/\text{min}$. t =pulse integration time interval. The resulting memory coefficients m are shown.

5. RESULTS.

The response of the detector to a pulse of radon is shown in Figure 2. In this case, a radon room, with uranium ore as emanator, is used as the radon source. Radon concentration in the room varies slowly in time due to meteorological conditions. To determine the radon concentration during the radon pulse, grab samples are taken from the room by pre-evacuated ZnS-cells. It is only the shortest time interval ($t = 5$ minutes) that can accurately resolve the radon pulse of 15 minutes duration. The uncorrected pulse count rate converted to activity units is shown for comparison. The lower part of Figure 2 illustrates how the measuring system corrects for a radon pulse shorter than the time interval over which the counts are integrated. In this case, the distortion of the square pulse input is expected, as the assumption of constant radon concentration in each time interval is invalidated.

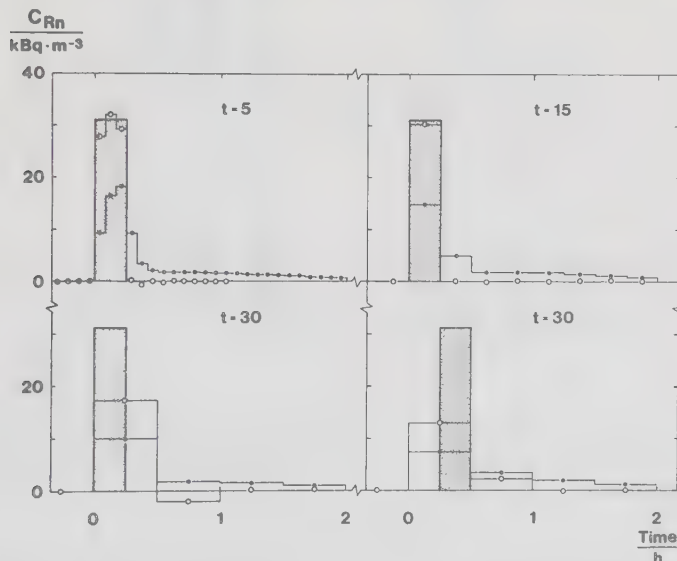


Figure 2. The response of the radon detector to a square peak of radon. The peak is 15 minutes long and the activity concentration is 31 kBq/m^3 . Air flow = $1.8 \text{ dm}^3/\text{min}$.

t = pulse integration time interval in minutes.

—●— = detector signal uncorrected;

—○— = detector signal corrected for the memory effect.

To investigate the system response to several rapid changes in radon concentration, air was sucked intermittently from a bag holding a radon concentration of 1075 Bq/m^3 . Before, in between and after the radon pulses the system was given radon-free air. The resulting uncorrected, and by the microprocessor corrected, radon concentration values are displayed in Figure 3. The short 'time constant' 5 minutes reproduces the true radon concentration well, except for the negative values after the first and third peak. The negative values are due to the finite wash-out time of radon gas, resulting in a positive m_2 -value, of Figure 1. These deficiencies in the theory become significant only in the shortest time interval, 5 minutes.

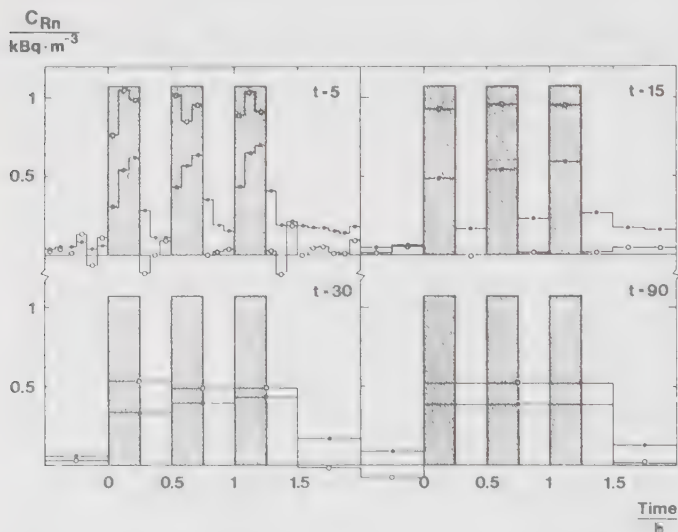


Figure 3. The response of the continuous radon detector to three radon peaks of 15 minutes duration each. The peak activity concentration is 1075 Bq/m^3 . The nomenclature is identical with that in Figure 2.

Investigations concerning the uncertainties in m , and thus the corrected radon values, are now being conducted. The corrected radon values when 15 minutes are used as time interval are close to the real peaks, but this is partly due to the coincidence between time interval and the radon pulse duration.

The continued work with the true radon detector in the near fu-

ture involves optimization of the light collection from the ZnS-cell and investigations to show how air flow and humidity affect the response.

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THE EFFECT OF AIR FILTRATION ON AEROSOL-ATTACHED
AND UNATTACHED ^{222}Rn DECAY PRODUCTS

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THE EFFECT OF AIR FILTRATION ON AEROSOL-ATTACHED
AND UNATTACHED ^{222}Rn DECAY PRODUCTS.

INTRODUCTION.

The accumulation of ^{222}Rn daughters in indoor air is a major source of natural background radiation in several countries (UN82). Regions with cold climates or seasons require energy-efficient dwellings with low air exchange rates. ^{222}Rn emanating from the ground or the building materials, is trapped in the house and the indoor activity concentration build-up can be considerable. If the soil gas infiltration is significant, high levels of radon can be reached even in well ventilated houses, but this is an exception to the rule and ventilation is normally an effective counter-measure. As straightforward ventilation wastes valuable energy, ventilation based on the reuse of air is preferred. The low, sometimes extremely low infiltration rates of energy-efficient houses necessitate air-cleaning methods. In addition air cleaners are common in dwellings where carpets, pet animals etc. cause allergy problems.

Room models have been published by Jacobi (Ja72) and Postendörfer et al (Po78) in studies how ventilation, and radon daughter plate-out etc. influence the indoor radon- and radon daughter concentration. Until recently experimental studies of ventilation- and air-treatment devices concerning indoor radon daughter levels have been limited to working-level measurements, with no distinction between attached and unattached radon daughter phases. During 1982-83 two research groups published papers covering the radiological aspects of indoor air cleaning, and making the necessary separation of activity attached and unattached to aerosol particles. Rudnik et al (Ru82, Hi83, Ru83) investigated two types, of fans and two types of air cleaners in a 78 m^3 chamber. They found a significantly reduced concentration of unattached radon daughters, attributable to the fans, due to plate-out on the surfaces of the chamber. The most efficient filter gave the least reduction in unattached activity (Hi83). No absorbed dose or dose equivalent calculations are presented by the Rudnik group. Jonassen and McLaughlin (Jo82) exploited air filtration using a fine filter in a large experimental room of volume 324 m^3 . They present their results as a relative absorbed dose to the basal cells of the bronchial epithelium, and they found significant dose reduction except for very low initial concentrations of aerosols (less than $100\text{--}500$ particles per cm^3). The filtration rate was varied up to about 4 h^{-1} . In the Harley-Pasternak dose model (Ha72), used by Jonassen and McLaughlin, contributions from the unattached RaB and RaC-C' activity are neglected.

The present experimental work was performed in the period July 1982- March 1983. The main objective of this investigation is to make an experimental assessment of how air filtration influences the airborne activity and the effective dose equivalent (ICRP77). In lung-dosimetry models the conversion factor for air activity concentration to absorbed dose in the respiratory tract, is about one order of magnitude greater for activity unattached to the aerosol, than the attached activity. By introducing an effective air filtration, the ratio of unattached to attached airborne activity increases significantly due to the net aerosol removal. Therefore, in any detailed study of the radiological effect of air filtration, a separation of unattached and attached activity is mandatory.

Apart from the relevance of air filtration in energy-efficient buildings there is also a potential remedial effect of air cleaning, especially in houses where increased ventilation has little effect, is unpractical or is too expensive.

Before proceeding to the air filtration results, a short description of the radon daughter behaviour in indoor air will be given, followed by a more detailed description of experimental methods and equipment used.

RADON DAUGHTERS IN INDOOR AIR.

In the following text radon is used to denote the isotope ^{222}Rn . Radon decays through a series of decay products. The first four of these are short-lived isotopes of heavy metals (Fig. 1). The very short-lived RaC' is always in equilibrium with RaC in the time scale relevant in this work. These two radionuclides can therefore be considered as one alpha- and beta-emitting nuclide. In illustrations below, this 'double' nuclide is designated RaC . Upon the decay of radon, RaA becomes a reactive positive ion. It has been assumed that RaA very rapidly combines with water vapour and trace gases in the air, forming particles still unattached to the aerosol (Bu81). These ultrafine particles, together with their charged or neutral atomic precursors are without further distinction both termed 'unattached' in the following text.

The airborne, unattached RaA in a room is eliminated primarily by radioactive decay, attachment to the ambient aerosol, deposition on macroscopic surfaces (plate-out and sedimentation) and ventilation. Human activity influences the eliminating paths, e.g. by changing ventilation or adding filtration through air-treatment devices (cf Fig. 1).

The attachment of radon daughters to an aerosol is a complicated process. The attachment probability depends on the charged state of both RaA and the aerosol as well as other physical and chemical properties of the unattached RaA and the aerosol particle. Several authors have studied the attachment to monodisperse aerosols and a survey by Ho et al (Ho82) contains references to the literature. Conflicting results have been reported, but it is obvious that the total available aerosol area, to a great extent, governs the attachment rate.

The recoil energy upon the alpha decay of RaA is large enough to detach RaB from an aerosol particle or a surface (Me76). This recoil effect adds further complexity to the airborne radon daughter 'budget' in a room. Both attached and unattached daughters are removed from the air by plate-out. The deposition rate depends on diffusivity, air turbulence, surface to volume ratio of the room and the physical and chemical properties of both the daughter particle and the surface. The high diffusion coefficient of unattached activity results in a plate-out rate, λ_p , of the order of 0.01 per second. The corresponding rate for the attached activity is considerably lower. Radon- and radon daughter (unattached and attached) inventory calculations, taking into account ventilation, attachment, plate-out, recoil and physical decay, have been performed (Po78, Br83).

EXPERIMENTAL METHODS AND UNCERTAINTIES.

The radon room.

A small radon room has been used in the cleaning experiments. The dimensions are 1.5x1.0x2.0 m³. The room is a closed duct in the concrete wall of the laboratory (Fig. 2). The walls are of thick concrete and the room is closed off from the nearby room by a 22 mm plywood wall. All inner surfaces are painted with plastic paint except the floor, which is covered by a fitted nylon carpet.

Several tubes and outlets have been installed in the wooden wall to permit air sampling and to provide electrical facilities in the room. Detectors, filter holders etc. can be introduced into the room through four large tubes of i.d. 160 mm. A small ceramic radium source (2 MBq, type RAE, Amersham, England) is connected to the room and a pump in a closed loop. Another air pump with variable flow ventilates the room. The inlet air is taken from the laboratory and the exhaust air is fed through a gas volume meter into the laboratory ventilation system.

A u-formed circular ventilation pipe is connected to the radon room. The pipe accommodates a filter holder and a fan. The speed of the fan is regulated by a variostat. The filters are $20 \times 20 \text{ cm}^2$ and thicknesses of up to 10 cm can be used. A differential pressure gauge is connected in parallel with the filter. To generate particles a heated filament of kanthal is used. The filament can be placed either inside the radon room or in a metal canister outside the room. The particles have, in the latter case, been pumped into the room.

The separation of unattached activity.

To separate the unattached daughter products from the attached ones, one can exploit the great difference in their diffusivity. The sideward movement of a particle per unit time due to collisions with gas molecules is proportional to the diffusion coefficient, modified by the Cunningham slip factor for small particles. If an air stream flows through a small tube, a high probability for the deposition of the unattached activity can be obtained, while the larger aerosol activity passes through the tube more or less unaffected. Recently van der Vooren et al surveyed the unattached radon daughter measurement techniques (Vo82).

In the present study passive wire screens of stainless steel were used to separate out the unattached radon daughter activity. To evaluate the collection efficiency for attached, a, and unattached, u, activity, three wire screens in series were backed up by a Millipore AA or FS filter. This technique was suggested to us by Jonassen (Jo81). To reconstruct the collection efficiencies, a and u, and the unattached, C_{ui} , and attached activity concentration, C_{ai} , from a measurement with the screen filter series, the following equations were set up (Sa82)

$$\begin{aligned} S_{ji} &= u(1-u)^{j-1}C_{ui} + a(1-a)^{j-1}C_{ai} & (\text{Bq/m}^3) \\ \text{Eq. 1} \quad F_i &= (1-u)^3C_{ui} + (1-a)^3C_{ai} & (-" -) \end{aligned}$$

where S_{ji} and F_i are the air concentration of the i :th species, corresponding to the measured activity on the j :th screen and the filter. The indices $i=1,2,3$ denote RaA, RaB and RaC respectively. Equation 1 is based on the assumptions that the backup filter has a 100 % collection efficiency for both unattached and attached activity and that the aerosol- and the unattached size distributions are not appreciably changed by passing through the screens. The latter assumption is valid if the efficiency, a, assumes a low value, and if the unattached activity is close to monodispersal. A little work on Equation 1 will give the

monotonously decreasing coefficients K

$$K_{1i} = C_{ui} + C_{ai} = S_{1i} + S_{2i} + S_{3i} + F_i$$

$$K_{2i} = uC_{ui} + aC_{ai} = S_{1i}$$

Eq. 2

$$K_{3i} = u^2 C_{ui} + a^2 C_{ai} = S_{1i} - S_{2i}$$

$$K_{4i} = u^3 C_{ui} + a^3 C_{ai} = S_{1i} - 2S_{2i} + S_{3i}$$

The analytical solutions for the efficiencies, a and u, are

$$\text{Eq. 3} \quad \begin{Bmatrix} u_i \\ a_i \end{Bmatrix} = \frac{K_{1i}K_{4i} - K_{2i}K_{3i}}{\underbrace{2(K_{1i}K_{3i} - K_{2i}^2)}_p} \begin{Bmatrix} + \\ - \end{Bmatrix} \sqrt{p^2 - \frac{K_{2i}K_{4i} - K_{3i}^2}{K_{1i}K_{3i} - K_{2i}^2}}$$

The screen-filter method is well suitable for the determination of u. Figure 3 shows the collection efficiency for unattached daughters, u, as a function of sampling velocity. The experimental results are compared with efficiencies calculated from the empirical equation of Thomas and Hinchliffe (Th72) and the theoretical formula of Cheng and collaborators (Ye82).

All calibration experiments point towards an efficiency, a, close to zero when equation 3 is used, but negative values and positive values as large as 5% can be encountered. This large spread in the collection efficiency, a, is caused by the subtraction in Eq. 3 of two nearly equal numbers. This low experimental accuracy in the aerosol collection efficiency, a, of the wire screen should be expected, when only three screens are used and the value of a is about 1% or less. We have performed theoretical calculations of the aerosol collection efficiency, a, according to the formula of Cheng. The results are given in Figures 4 and 5. The filament produces relatively small aerosol particles compared with a typical indoor aerosol (cf Fig. 18 below). Activity median diameters (AMD) of about 0.06 μm and a geometrical standard deviation, $\sigma_g=2$ correspond to $a=1\%$ in Figure 5. This value of a has been used in all calculations presented in this report. Typical indoor aerosols have area medians in the interval 0.1-0.3 μm and $\sigma_g=2$ (Jo83). Postendörfer et al characterize the typical indoor aerosol by an AMD in the range from 0.1 to 0.2 μm (Po78).

The counting efficiency.

The activity of the filter and wire screens was measured in vacuum using surface-barrier detectors. The number of counts in the alpha peaks for RaA and RaC', at two different time intervals after sampling, were recorded in a multi-channel analyser. From these numbers (corrected for background and contamination from previous counting periods) the air concentrations of RaA, RaB and RaC are calculated by a Fortran program on a PDP11/40 computer.

The counting efficiency of a detector for alpha activity on a wire screen deserves some attention. The exact activity distribution around the cylindrical wires of the screen depends on the sampling velocity. For the wire screen used, the dependence is significant only for velocities of less than 10 cm s^{-1} (Figure 6). For the same sampled activity the counting efficiency is lower for the wire screen compared with a filter. The wires of the screen conceal some of the alpha activity from the detector. To quantify this phenomenon simultaneous gamma- and alpha measurements on filter and screens were made. For this purpose a small diffused-junction alpha detector was inserted into a NaI well-crystal gamma detector. Figure 7 shows a spectrum from one of these measurements. Assuming the same gamma counting efficiency for screen and filter gives an alpha counting efficiency 20% lower for a screen compared to a filter, for sampling velocities in the range $10\text{-}20 \text{ cm s}^{-1}$.

Activity evaluation.

In the air-cleaning experiments a filter holder accomodating one wire screen in front of a membrane filter was used. The experimental efficiency, u , in Figure 3 was parametrized and included in a computer program. The input parameters are the sampling velocity, the efficiency a , and the activity concentrations of RaA, RaB and RaC, on wire screen and filter. The program calculates the potential alpha energy concentrations of the individual daughters for attached and unattached activity, the fraction of unattached potential alpha energy, f_p , and the effective dose equivalent rate according to different dose models.

To measure the radon concentration prefiltered air from the room is continuously drawn through a small ZnS flask attached to a PM-tube. After correction for detector background, the alpha count-rate is converted to radon concentration units. The ZnS flask has a volume of 0.16 litres and is of our own construction. The dynamic sensitivity, i.e. during through flow conditions, is about 1.6 counts per second per Bq. Due to plate-out of radon decay products on the inside surfaces of the flask the radon signal is impaired by a 'memory' effect. This effect is of importance only for short-time variations and has not been corrected for in this work.

Aerosol equipment.

The aerosol concentration, (denoted by CNC or N in the Figures), in the radon room was continuously registered by a condensation nucleus counter (TSI 3030 CNC, St. Paul, Minnesota, USA). The loss of particles in the tube connecting the condensation nuclei counter (CNC) to the radon room is of the order of a few percent. The exact value depends on the aerosol size distribution. No dead-time correction to the CNC values has been applied. The dead-time correction has a maximum of 20% and is relevant only in the single particle counting regime, i.e. <1000 particles/cm³.

In most of the air-cleaning experiments a particle size analyser (TSI 3030 EAA, St. Paul, Minnesota, USA) was used. When this was used the aerosol inlet of the analyser was connected to the radon room through the same tube as the CN counter. The sheet air (46 l min^{-1}) to the analyser was taken from the laboratory outside the radon room. The analyser current output is used in a computer program, to evaluate the number-, surface- and volume distribution of the aerosol (Joh82).

Effective dose equivalent.

Several dose models connecting airborne radon daughter activity and the absorbed dose in the respiratory airways have been published. In this work the recent models of Jacobi and Eisfeld (J-E), and James and Birchall (J-B), as described in ICRP report 32 (ICRP81), are used. The effective whole-body dose equivalent rate, $\dot{H}$, is calculated by

$$\begin{array}{lll} \dot{H}/\dot{I}_{\text{pot}} = 1.39 + 9.9 f_p & (\text{Sv J}^{-1}) & \text{J-E dose model} \\ \text{Eq. 4} & & \\ \dot{H}/\dot{I}_{\text{pot}} = 0.96 + 34 f_p & (\text{Sv J}^{-1}) & \text{J-B dose model} \end{array}$$

where $\dot{I}_{\text{pot}}$ (J h^{-1}) is the inhalation rate of the total potential alpha energy and f_p is the fraction of the total potential alpha energy unattached to the aerosol. Equation 4 is based on the regional lung dose concept with a weighting factor of $w_T=0.06$ for both the trachea-bronchial and the pulmonary regions (ICRP81). The contributions to the effective dose equivalent from radon daughters in the bronchial epithelium and the alveolar region are combined, and the dose equivalent from radon itself is neglected. Our computer program calculation of $\dot{I}_{\text{pot}}$, rests on the assumption of a respiration rate of $1.2 \text{ m}^3 \text{ h}^{-1}$. Equation 4 is valid for an activity median diameter in the range $0.2\text{--}0.3 \text{ }\mu\text{m}$.

Uncertainties in the radon daughter concentrations.

The error in the individual determinations of the radon daughter concentrations is of a systematic and statistical nature. The systematic error has not been evaluated, as relative concentrations of the daughters before and after the implementation of air filtration give sufficient knowledge. The radon concentration presented can be directly traced back to standard radium solutions from the National Bureau of Standards, USA, while the radon daughter activities are obtained by calibrating the surface barrier detector with plated alpha sources. This latter calibration method should not be considered as an absolute one, as long as the differences between the plated- and filter sources in self-absorption, surface homogeneity of activity etc. are not thoroughly investigated. From the results shown in the Figures below it is possible to extract equilibrium factors between the daughters and radon. These equilibrium factors may be in error, due to the non-absolute calibration of the daughter activity. Calibration procedures now underway indicate that the radon daughter activities, and therefore also the equilibrium factors, presented in this investigation are too small. This is expected, due to the self-absorption of alpha particles in the filter. So in conclusion, the radon daughter values and the potential alpha energy equilibrium factors should only be judged relative to each other.

The stochastic error in the radon daughter activities has been evaluated in the following way. In the first air-cleaning experiments three to six air samples were taken on the same day and with the same filtering-, aerosol-, ventilation- etc. arrangements. These repetitive measurements have been statistically analysed and the relative standard deviation calculated (Figure 8). Concentrations below 20 Bq/m³ and results from one day, when the aerosol concentration changed by more than 50%, have been excluded. The results in Figure 8 were obtained from an air sample of about 50 l, a wire screen collection efficiency for unattached daughters of ~70%, a detector alpha counting efficiency of ~20% and two counting time intervals 1-11 and 30-50 minutes after the cessation of the air sampling. As a complement to the experimental uncertainties in Figure 8 a theoretical estimation of the error in the RaA concentration was performed. The RaA concentration can be written (sampling starts at t=0):

$$\text{Eq. 5} \quad C_A = \frac{(P - P_b) \cdot \lambda_A}{V \cdot \epsilon \cdot \eta \cdot E(-t_s) \cdot I(t_s) [E(t_1) - E(t_2)]} \quad (\text{Bq m}^{-3})$$

where P is the number of pulses in the RaA-peak
 P_b is the number of background pulses in the RaA-peak.
 t_s is the sampling time
 $t_2 - t_1$ is the pulse accumulating time interval
 λ_A is the decay constant of RaA
 $E(t) = \exp(-\lambda_A t)$
 $I(t) = (1 - \exp\{-\lambda_A \cdot t\}) / \lambda_A$
 V air-sampling rate
 ϵ counting efficiency
 η collection efficiency of filter/wire screen.

A sampling time of 600 s and the above cited parameter values inserted in Equation 5 give

$$C_A \cong 2 \cdot 10^{-5} \frac{P - P_b}{V \cdot \epsilon \cdot \eta} = 2 \cdot 10^{-5} \frac{P_n}{V \cdot \epsilon \cdot \eta}$$

If we assume that only variations in pulse counts, background, sampling rate and efficiencies contribute, we have

$$\text{Eq. 6} \quad \left(\frac{s_C}{C_A} \right)^2 = \left(\frac{s_P}{P_n} \right)^2 + \left(\frac{s_V}{V} \right)^2 + \left(\frac{s_\epsilon}{\epsilon} \right)^2 + \left(\frac{s_\eta}{\eta} \right)^2$$

where s_i^2 is the estimated variance in parameter i . The background pulse count rate from a filter or wire screen in a surface barrier detector is very low. Contamination increases the background, when radiation from filters is measured in vacuum, but mainly in the RaC' alpha peak. The background in the RaA peak region covering the alpha energy interval 4.0-6.2 MeV, is typically 0.1 cpm, but can occasionally reach 1-2 cpm after heavy usage of the detector.

The experimental values in Figure 8 include variations in the individual radon daughter concentrations, during an experimental day, caused by undeliberate changes in the radon level, aerosol number and size distribution etc. It is therefore reasonable to compare Equation 6 with experimental values in Figure 8 somewhat lower than the curve actually drawn. If this is done at the high concentration end, where the relative variance in pulse count is small, it is concluded that

$$\left(\frac{s_V}{V} \right)^2 + \left(\frac{s_\epsilon}{\epsilon} \right)^2 + \left(\frac{s_\eta}{\eta} \right)^2 \approx 0.05^2$$

and thus we have

$$\text{Eq. 7} \quad \left(\frac{s_C}{C_A} \right) = 100 \left[(P_n + 2P_b)/P_n^2 + 0.05^2 \right]^{\frac{1}{2}} \quad (\%)$$

if the pulse statistics follow a Poisson distribution. Calculations according to Equation 7, for two different background count rates, are plotted in Figure 8 as 'theoretical' values.

None of the individual daughters nor phases (attached or unattached) show any systematic digression from the general trend in Figure 8. It seems that the alpha spectrometry evaluation method outlined above results in approximately the same accuracy for all three daughters in both phases, at identical air concentrations.

RESULTS AND DISCUSSION.

The idea for the present work dates back to the autumn of 1979 when we used different air cleaners in a 'radon' house. The radon concentration and the filterable alpha concentration in air were continuously recorded. The air cleaners had a capacity in the range 100-150 m³ h⁻¹. A significant decrease in the filterable alpha concentration was clearly indicated. Conclusions about any dose reducing effects of the air cleaning could not be extracted, as a separation of individual daughter and unattached/attached daughter activity was not at hand. Therefore a preliminary experimental series, separating attached and unattached activity in the dwelling was performed. The results of these experiments are shown in Figure 9. The filter holder accommodated a wire screen of diameter 8.0 cm in front of a Whatman GF/A filter. The activity on the filters was evaluated giving an EER-value (Equivalent Equilibrium Radon concentration, Kusnetz method), and the wire screen activity was measured according to the modified Tsivoglou technique (IAEA76). The latter separates the individual daughter concentrations. The air-cleaning rate, AC, in Figure 9 is expressed as the number of total room volumes filtered per hour. The total room volume was defined as the volume of the rooms connected to the air-cleaner room by open doors.

The largest concentration of unattached RaA is experienced at an air flow rate of 8 room volumes per hour. The question was raised, of whether air filtering actually increases the absolute value of the unattached activity concentration.

Figure 9 gives no definite answer as air sampling took place over a period of four days. Other factors beyond the air cleaner could easily have been the source of the high concentration of unattached RaA. Therefore it was decided to perform air-cleaning investigations in the laboratory where the surrounding conditions could be better controlled. Unfortunately, other obligations interrupted the work and the laboratory experiments had to be postponed until 1982.

In a series of experiments in the radon cell (cf Fig. 2) different filter media were tested. Most of the filters are classified as 'fine' and are commercially available, cf Table 1. Electret filters are denoted by an 'e' in the table.

TABLE 1.

Symbol	Filter	Type	Thickness (mm)	Surface weight (g/cm ⁻²)	Manufacturer/Supplier
G	G1300	Fine, e	5	3.1	Verto, Holland
C	CM285	Fine	5	0.6	Camfil, Sweden
W	4401	Wire-screen	0.1	-	Derma, Sweden
F4	F45	Fine	15	2.8	Carma, Sweden
F9	F95	Fine, e	5	2.5	Carma, Sweden
G8	G85	Fine	15	3.2	Carma, Sweden
A	CM115	Micro	0.8	0.9	Camfil, Sweden
P	peat	-	25	21	-

The results of two experimental series are given in Figure 10. The air exchange rate in the room is kept at 0.25 room volumes per hour, the temperature 22 ± 1 °C and the relative humidity in the interval 50-60%. No aerosol was generated in the radon room during the first series. The aerosol particles still present originate mainly from the laboratory air, which is fed into the room by the ventilation pump. The radon daughter concentrations are displayed as mean values of 3-5 samples. No corrections were made for the variations in radon level. The aerosol analyser was not available during the series shown in Figure 10a-f (the letters a, b etc. refer to the different experimental runs shown).

The low initial aerosol concentration results in a low equilibrium factor, and a relatively high fraction of unattached potential alpha energy ($f_p = 0.15$) in run 10a. Merely agitating the air using the air cleaner fan, as in experiments 10b and 10c, reduces the attached activity significantly. The effect on the unattached activity is more complicated. There is a decrease in RaA-u at $AC = 5 \text{ h}^{-1}$

but this reduction can also be attributed to the lowered radon level and the higher CN concentration. Agitating the air will increase the plate-out of unattached activity on all surfaces in the room and in the air-cleaning system, but variations in the aerosol concentration and size distribution can easily conceal this higher plate-out trend, as the attachment rate of the unattached activity to the aerosol is changed.

Filtering the air at a rate of 2 h^{-1} (run 10d) increases the concentration of unattached RaA. The increase is $\sim 15\%$, comparing 10a and 10d and correcting for the decreasing radon concentration. The 100% filter efficiency for unattached activity is more than compensated for by the drop in attachment rate due to lowered aerosol concentration. At the higher circulation rates, 5 and 13 h^{-1} , a net filtering effect for RaA-u is noticed (cf runs 10d-f).

The low aerosol load in runs 10a-f means that a large fraction of the RaA activity is in the unattached mode. The dose models of J-E and J-B have quite different weighting factors for the unattached activity (cf Eq. 2). This results in discrepancies in the calculated effective dose equivalents for the two models. Initially, with the air cleaner off (Fig. 10a), f_p is 0.15, and the J-B model shows an effective dose equivalent rate 100% greater than the J-E model. At higher filtering rates, f_p is even higher, and the dose equivalents disagree by a factor of ~ 3 . It should be observed however that both models predict a net positive effect at the circulation rates 5 and 13 room volumes per hour. The effective dose equivalents at 13 h^{-1} (Fig. 10f), adjusted for the decrease in radon level, are 66% (J-B model) and 44% (J-E model) of the initial value without the air cleaner (Fig. 10a).

After the experiments in 10a-f the aerosol concentration was raised by heating the filament inside the radon room. The particle analyser (TSI 3030) was now added to the equipment arsenal, to give the aerosol size distribution. In Figures 10g-k the radioactivity results are given, and Figure 11 shows the particle number size distribution. The undisturbed aerosol (10g) exhibits approximately a log-normal distribution. The number and surface median are 15 and 40 nm with a geometric standard deviation, σ_g , equal to about 2.0 for both distributions. The analyser does not measure aerosol particles of diameters less than 10 nm. Also the sensitivity of the CN counter drops if the particle diameter falls below 20 nm. In Figure 10g the analyser gives the total number of particles larger than 10 nm as 290 mm^{-3} while the CN counter shows a slightly higher value, 320 mm^{-3} .

The small-sized aerosol increases the radon equilibrium factor from 0.22 (10a) to 0.30 (10g). The fraction of unattached potential alpha energy, f_p , is drastically reduced to 0.016 (10g) due to the higher aerosol load. Comparing 10g with 10 l-m, the air cleaner with filter G or C reduces the total aerosol surface by almost a factor of ten. And the activity response to this is a dramatic decrease in attached activity and a moderate increase ($\sim 25\%$) in unattached RaA. By filtering at the rate of 13 h^{-1} the unattached potential alpha energy fraction, f_p , is raised from 0.016 to 0.16. The reduction in effective dose equivalent is large, a factor of ~ 2 or ~ 4 , depending on dose model, and taking into consideration the slightly increasing radon level during 10g-j. Agitating the air is less effective at the high aerosol concentration than at low aerosol load (cf 10g-h and 10a-b).

In 10k a wire screen (aperture 0.09 and wire diameter 0.056 mm) replaced the filter in the air cleaner. The circulation rate of 7.7 h^{-1} equals an air velocity of 0.30 m/s . The collection efficiency of the wire screen for unattached activity is around 80% at this velocity, while the corresponding figure for attached activity is $\sim 1\%$. It is obvious that filtration, at a rate 8 h^{-1} , of the unattached activity exclusively has only a limited effect on the dose equivalent. The effective filtration rate of the unattached activity in the wire screen, ($\sim 6 \text{ h}^{-1}$ in 10k) can not match the fast elimination by attachment to the aerosol.

The experimental series was rounded off by a measurement without the air cleaner or aerosol production (10l), to check the reproducibility of the radon room and sampling/detection routine. The values in 10l (and also 10k) are mean values of only two air samples, and thus slightly more uncertain than the other results in Figure 10 (4-5 samples). The time lag between 10a and 10l is 11 days. Ignoring the unattached RaB and RaC, all activities could be reproduced within 10%, if a correction for the difference in radon concentration is made.

The air exchange rate of 13 h^{-1} in the air-cleaning filter is unpractically high for use in dwellings. In an ordinary room of volume 50 m^3 it would correspond to a rate of $650 \text{ m}^3 \text{ h}^{-1}$, creating noise problems and a high energy consumption. A logical follow-up to this series of experiments was therefore a series of filter experiments at lower air-exchange rates. To improve the accuracy at the low circulation rates a gas meter was installed in the radon room, in series with and behind the filter and air-cleaning fan. Due to the inertia of the shovel of the gas meter the exchange rate had to be greater than or equal to 2 h^{-1} to be readable. An experimental series covering about a week was initiated to evaluate the performance of filter CM 285 at a low air-filtering rate. The particle size

distribution was found to change during the experiments. These aerosol changes lead to our obtaining more information from the experiments than initially intended and provides further justification for a separate depiction of the results. The activity concentrations are shown in Figure 12, consecutively from left to right. As before each experimental point is from one and the same day and is a mean value of 3-4 air samples, with the exception of run 12f, which is a single sample. On every experimental day two runs, separated by several hours, were carried out with the particle analyser (Figure 13). The disadvantage of having an unknown number of very small particles (cf Figure 10), was now partially avoided by putting the glowing filament into a closed canister outside the radon room, and pumping the aerosol particles through a tube into the room. The very small particles from the filament have a low probability of reaching the radon room, due to an effective plate-out on the inner walls of the tube.

Initially in 12a the CN count is much less than in 10g. The values of the total surface area of the aerosol, as given by the analyser, are $1200 \text{ and } 600 \mu\text{m}^2 \text{ cm}^{-3}$ in 10g and 12a respectively, explaining the only moderately lower aerosol activity in 12a. On the next sampling day (run 12b) the CN count had dropped to 21 mm^{-3} but the trend of increasing attached activity and decreasing RaA-u is not the expected. The subsequent computer evaluation of the analyser signal gave the explanation. In 12b the number size distribution changed towards larger particles. As the CN count decreased from 40 to 21 mm^{-3} between 12a and b, the total aerosol surface increased from 600 to $1600 \mu\text{m}^2 \text{ cm}^{-3}$. Cleaning the air at 3.4 h^{-1} introduces a drastically new distribution of activity between attached and unattached phases. But this dramatic change is a combined effect of the filtration implementation and a changed aerosol size distribution, (cf Fig. 13).

Comparing the results in the cases air cleaner off/on at 3.4 h^{-1} (see 12c and 12e), and assuming no aerosol changes in between, the reduction in effective dose equivalent by air cleaning is 20% (J-B) and 30% (J-E). The correction for the deviation in radon concentration has been applied. Later tests of filter C confirmed that this dose equivalent reduction could be expected for the parameters at hand. The results in Figure 12, underline the necessity of controlling the aerosol stability during air-cleaning experiments.

In Figure 12f the aerosol production was increased to give about the same number of particles as initially in 12a. The particles in 12f are much smaller however and the total surface area of the aerosol is only 30% of that in 12a. The resulting lower attachment rate then explains the observed increase in unattached RaA (RaA-u) and decrease in aerosol daughter activity.

Results and calculations from air-cleaning experiments at filtering rates of 5 h^{-1} and 9 h^{-1} are shown in Figures 14 and 15. Single sampling values are displayed as a function of time. In many cases the exchange of filters is so close in time that equilibrium conditions are not reached. When this is the case, the results can not be considered as a test of the filter used. As before the aerosol source was the heated filament in the canister outside the radon room (the filament was not used during 23/12 Fig. 15).

In spite of the non-equilibrium conditions it is obvious that just circulating the air through the air cleaner without a filter, significantly lowers the effective dose equivalent. It should be remembered that the air-cleaning system, besides the ventilation channel and fan, (cf Fig. 2) involves a connecting plastic tube of length 1.0 m to the gas volume meter. Furthermore the output of the gasmeter is fed to the interior of the radon room through another plastic tube of length 1.0 m. The small-sized aerosol, typically a surface area median of 100 nm and $\sigma_g=2.0$, is, to a great extent, lost in the ventilation system during air circulation even without a filter in place. In addition there will be a greater plate-out onto the walls of the radon room, due to turbulence and better mixing, when the fan is working. An air cleaning at rates of 5 and 9 h^{-1} increases the unattached fraction f_p considerably. The maximum f_p -value reached by the electret filter F9 with a circulation rate 9 h^{-1} is 0.80. On three occasions the air filtration was left off overnight. Comparing the results of the first sampling the next morning, with air filtration off, illustrates the reproducibility of the "climate" in the radon room and the results obtained with this measuring technique. The effective dose equivalent rates recover to within 15% in the J-B model and 5% in the J-E model. The higher value for the J-B dose model reflects its higher sensitivity to the variative unattached activity, dominated by RaA-u.

An absolute and a fine filter combined gave a high air resistance. A maximum rate of only 2.3 h^{-1} could be achieved (Fig. 14 G8+A). The filter combination G8+A at 2.3 h^{-1} and the fine filter G8 at 5.6 h^{-1} removed the attached activity at about the same rate, due to the lower filtration efficiency of the latter. But as the RaA-u is removed more effectively by G8 at 5.6 h^{-1} , this combination is the better one from the dosimetric point of view.

The effect of air cleaning on the unattached RaA and the attached activity, at an exchange rate of 9 h^{-1} , is demonstrated in Figure 15. A net filtering effect is seen for all daughters and phases. At this high air-cleaning rate the dose equivalent reduction is quite clear. When corrected for differences in radon level the dose equivalent rate is reduced by a factor of ~ 3 (model J-B) or ~ 4.5

(J-E) by the electret filter F9 at $\sim 9 \text{ h}^{-1}$.

Two commercial air cleaners were also tested in the radon room. Both cleaners are intended for use in rooms of a dwelling. The Remington AP-100 is a small table model fitted with an electret filter. The Ventronic Braun, type ACL, is a portable floor model containing an electro-filter preceded by a fine filter (CM 285). According to the manual the two speeds of the Braun cleaner correspond to 110 and $50 \text{ m}^3/\text{h}$. The air-cleaning capacity of the Remington AP-100 is not given in its manual.

When tested the air cleaners were placed on the floor of the radon room. The higher speed of the Braun cleaner was used. At this rating the air volume exchange rate in the 3 m^3 radon room is extremely high, $\sim 36 \text{ h}^{-1}$. The results with the air cleaners off and on are displayed in Figures 16 and 17. The radon concentration was within $\sim 5\%$ during the tests. The radioactivity levels are plotted down to 1 Bq m^{-3} , but only values above 50 Bq m^{-3} are accurate enough to be seriously considered (cf Fig. 9). The condensation nuclei concentration is displayed in a linear scale as copied from the recorder output of the CN counter. The slowly decaying CN concentration with the air cleaners off, also noted on other occasions, is due to non-uniformity in the aerosol emission rate of the filament.

The Remington AP-100 reduces the dose equivalent by a factor of 2-3, depending on dose model, in the 3 m^3 room. Assuming a constant aerosol source strength and cleaning rate of 36 h^{-1} for the Braun cleaner (see Fig. 17) gives an effective aerosol filtering rate of $2-3 \text{ h}^{-1}$ for the the AP-100. When these results are converted to most dwelling situations the influence of the Remington AP-100 on the effective dose equivalent rates would be small. At the high exchange rate of the Braun cleaner, the effective dose equivalent rate is completely dominated by the RaA-u component. The unattached fraction of potential alpha energy, f_p , is 0.88 and the dose equivalent reduction factor obtained is about 4 and 5 in the (J-B) and (J-E) dose models respectively.

Effect of air filtration on the activity median diameter (AMD).

The fraction of unattached potential activity, f_p , is greatly increased by air filtration, as shown in the experiments above. In the theoretical lung dose models used, the weighting factor for unattached activity is high, between ~ 10 and ~ 30 . Thus, the determination of f_p must have a high priority in every air filtration study. Another property of the radon daughters, influencing the dose equivalent calculation, is the size distribution of the aerosol activity. Log-normal distributions are unambiguously defined by their median diameter and the

geometric standard deviation σ_g . All dose equivalent calculations in this paper assume an activity median diameter (AMD) in the range 0.2-0.3 μm and $\sigma_g \approx 2$. As the AMDs in this investigation have been about 0.1 μm and below, the effective dose equivalent rates calculated should be considered as relative values. The effective dose equivalent rate will increase with decreasing AMD in the AMD-range 0.1-0.3 μm (Ja81). The exact relationship between AMD and effective dose equivalent rate is complicated and differs from one dose model to another. Roughly however, the effective dose equivalent rate is inversely proportional to the AMD in the AMD-interval mentioned.

As the dose model calculations are applied both with air cleaners on and off it is of interest to investigate how the AMD and σ_g are changed by air cleaning. Changes in the AMD are relatively more important in the J-E model, with its less pronounced f_p -dependence (cf Eq. 2). A direct measurement of the activity size distribution, during an effective air filtration is difficult, due to the low activity concentrations involved. An approximate and indirect way to assess the effect of air cleaning on the AMD is by looking at the aerosol surface area distribution.

The attachment coefficient of radon daughters is dependent on the size of the aerosol particle. An attachment coefficient proportional to particle radius, r , radius squared, r^2 , or something between r and r^2 , can be found in the review paper by Ho et al (Ho82). If for example the attachment coefficient goes as r^2 , the surface area and the activity size distribution will be identical. If the implementation of air filtration changes the surface area distribution towards larger particles, the activity size distribution will change in the same direction, if the attachment process as such is unaffected by the filtration. Some information is therefore gained by studying the effect of air filtration on the aerosol surface area distribution, a distribution obtained from the particle size analyser (EEA).

Figure 18 shows log-normal plots of the cumulative aerosol surface area during cleaning and non-cleaning situations. Figure 18a reveals that the air cleaner without a filter, or the air cleaner with a wire screen, has no effect on the surface area distribution. The hatched regions in Figure 18 correspond to the distributions with air cleaner off (two measurements). During highly efficient air filtration, such as the case using filter F9 in Figure 18b, the experimental uncertainty is considerable due to the small total aerosol area involved. The results in Figures 18c and d involve repetitive measurements, thereby giving some idea of the reproducibility. According to the distributions in Figures 18b-

d it is obvious that the relative surface distribution, if changed at all, reveals a tendency towards larger particles. Thus indicating an increase in the AMD, and therefore a decrease in the effective dose equivalent rate, during air cleaning. It must be stressed that this increase of the AMD may not be generally applicable to air cleaning, but could be expected when the median surface of the initial aerosol distribution is below the penetration 'window' of the air-cleaning filter.

CONCLUSIONS.

The results above verify that a sampling system consisting of a wire screen and a back-up filter, combined with alpha spectrometry measurements provides a convenient technique for separating attached and unattached radon daughters. To achieve high precision however the wire screen and filter must be carefully calibrated relative to each other for the particular air flow and aerosol used. When the unattached fraction of radon daughters is small it may be necessary to consider the collection efficiency of the wire screen for activity attached to the aerosol. Concerning the precision it is concluded that the standard deviation in individual daughter activities is 10 % or better for activity concentrations $> 100 \text{ Bq m}^{-3}$, if a 50 l sample is taken.

The experiments confirm that agitation and filtration of the air can have a considerable effect on the airborne radon daughter concentrations. Through filtration the unattached fraction of potential alpha energy increases considerably. The increase of f_p during air filtration is the main concern from the dosimetric point of view. The change in the AMD by air filtration is of much less importance. The exact effect of an air cleaner depends, to a large extent, on the initial aerosol load and surface area distribution. Figure 19 shows the relative reduction in dose equivalent rate at different filtration rates. To achieve a significant dose equivalent reduction a filtration rate of 5 h^{-1} or higher should be used. If initially clean conditions (normally not met in dwellings) are combined with low air filtration rates, the existence of an effective dose equivalent reduction can be questioned in dose models with a high weighting factor for the unattached activity. On the other hand, no situation in dwellings can be foreseen where the installation of an air cleaner will increase the effective dose equivalent significantly. There is either no effect at all or a reduction in the effective dose equivalent.

The complexity of the radon daughter elimination in room air makes straightforward judgements on the air-cleaning issue difficult. Further investigations are

clearly indicated in the search for the optimum air filtration or treatment conditions.

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FIGURE CAPTIONS.

1. A compartment model, illustrating the different phases and flow routes of radon and decay products. Ventilation (vent.) and filtration (filtr.) may remove airborne phases of the daughters. Decay data are given in the right part of the figure.
2. A horizontal cross section of the radon cell (schematic).
FH= Filter Holder for the cleaning filter.
3. The wire screen collection efficiency, u , for unattached radon daughters, as a function of air sampling velocity, v . The aperture and wire diameter are 0.25 mm and 0.16 mm respectively (24 cm^{-1}). The straight line is used in the computer calculations in this report.
 - theory, formula of Cheng (Ye82). Diffusion coefficient
 $= 0.054 \text{ cm}^2 \text{ s}^{-1}$.
 - Thomas-Hinchliffe semi-empirical formula (Th72).
 - + experiments in this work.
4. Theoretical calculation of the collection efficiency for aerosol particles of different diameters, according to the formula of Cheng (Ye82), for two different sampling velocities and for a wire screen of mesh 24 cm^{-1} .
5. Identical to Figure 4, but for log-normal size distributions of different medians and geometrical standard deviations, σ_g . The curves are the same as long as the collection efficiency refers to the same aerosol parameter as the median, e.g. activity.
6. The fraction of the ^{214}Po (RaC') alpha particles emitted (and detected) from the front of the wire screen. The number of alpha particles detected from back plus front is the total number. The data for the wire screen of mesh 79 cm^{-1} , are taken from James et al (Jam72).
7. Simultaneous alpha- and gamma spectra obtained from a small wire screen (24 cm^{-1}). The alpha detector is a small diffused-junction detector, placed inside a NaI(Tl) well crystal. Air sampling time = 10 minutes.
 - a and b: 60-360 seconds after sampling
 - c and d: 1800-2400 -----"-----

8. Experimental precision (relative standard deviation, $\sigma = s_C/C_A$) as a function of activity concentration. The air volume sampled is about 50 litres (se text). The full line is a fit by eye to the experimental points.

u= unattached, a= attached activity.

□ RaA-u ■ RaA-a

○ RaB-u ● RaB-a

△ RaC-u ▲ RaC-a

--◇-- Eq. 7, RaA-background= 0.1 cpm (se text)

—★— Eq. 7, —" = 1 —"

9. Air cleaning results in a dwelling. The air cleaner is of a box type with a fan inside. The filter material consists of two 5 cm thick glass-wool sheets normally used as insulating material. The air capacity is about 150 m³ h⁻¹.

AC/h⁻¹ Air cleaning rate per hour.

■ RaA-unattached.

○ RaB-unattached.

△ RaC-unattached.

★ Attached (Kusnetz).

10. The unattached (-u) and attached (-a) activity concentrations in the radon room for different combinations of filter, air-filtration rate (room volumes per hour), and aerosol load. In the heading are given the effective dose equivalent rates for the dose models of James-Birchall (H_{JB}) and Jacobi-Eisfeld (H_{JE}). For details, see text.

—x— radon

--□-- RaA-u —■— RaA-a

--△-- RaB-u —▲— RaA-a

--○-- RaC-u —●— RaC-a

11. Examples of the particle size distributions, obtained during the measurements illustrated in Figure 10. The number concentration of particles per logarithmic size interval, ϕ (dN/d ϕ), is plotted as a function of particle diameter, ϕ .

▲ Nov. 23 AC/off Filter/- (Fig. 10g)

● Nov. 24 AC/13 h⁻¹ Filter/- (Fig. 10h)

○ Nov. 26 AC/13 h⁻¹ Filter/C (Fig. 10j)

△ Nov. 26 AC/8 h⁻¹ Filter/W (Fig. 10k)

12. Air-cleaning results in the radon room. The notations are identical to those of Figure 10.

13. Aerosol size distributions obtained during the measurements shown in Figure 12 (cf Fig.11).

14. The activity (A) and aerosol particle (N) concentrations (upper part), and the corresponding effective dose equivalent rates (H) and unattached fractions of potential alpha energy (f_p), (lower part). The filter code is given in Table 1. The pressure drop (ΔP) over the filter, and the air-cleaning rates (AC) are also displayed.

- + James-Birchall dose model (J-B)
- o Jacobi-Eisfeld dose model (J-E)

15. As in Figure 14, but for other air-cleaning rates and filters.

16. Air-cleaning results in the radon room for the small table cleaner Remington AP-100. The scale for unattached (-u) and attached (-a) activity is to the left. The CN counter values, N, (scale to the right) are in ten thousands per cm^3 .

17. The same as in Figure 16, but for the Braun ACL cleaner.

18. Log-normal plots of the aerosol surface area distributions, during cleaning and non-cleaning situations. AC= air-filtration rate in room volumes per hour. Hatched regions = air cleaner off.

- a. ● Filter/- AC/13 h^{-1}
 + Filter/W AC/7.7 h^{-1}
- b. ● Filter/F4 AC/5.1 h^{-1}
 + Filter/F9 AC/8.7 h^{-1}
- c. ● Filter/G AC/5.6 h^{-1}
 + Filter/G AC/9.2 h^{-1}
- d. ○ Cleaner off (before cleaning).
 + Air cleaner AP-100 on.
 Δ Cleaner off (after cleaning).
 (cf Figure 16.)

19. The relative effective dose equivalent as a function of the filtration rate, AC, expressed in room volumes per hour. CN = initial condensation nuclei concentration (mm^{-3}).

Symbol	Filter	AC	CN
□	G	2.0	2.4
◇	G8+A	2.3	34
○	C	3.4	10
□	G	5.0	2.4
▽	F9	5.0	36
◇	P	5.1	34
▽	F4	5.1	36
◇	G8	5.6	34
+	F9	8.7	45
△	G,C	13.0	325

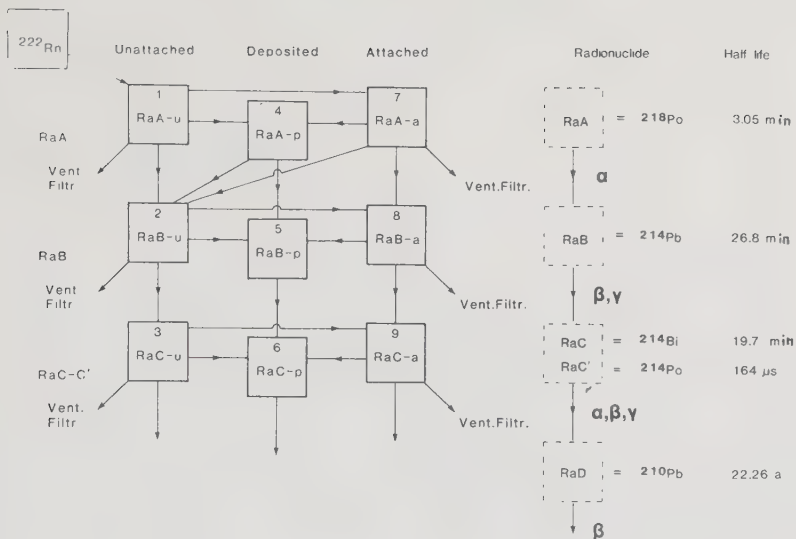


Figure 1.

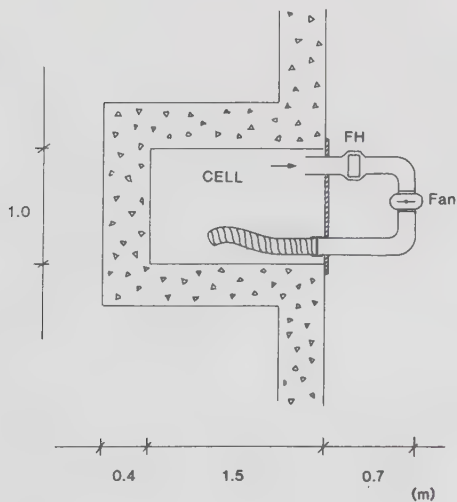


Figure 2.

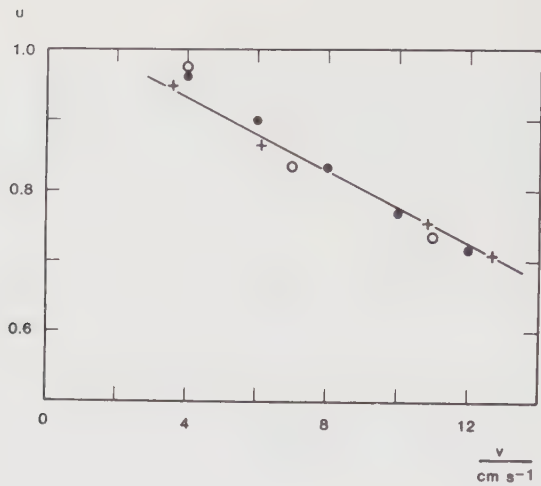


Figure 3.

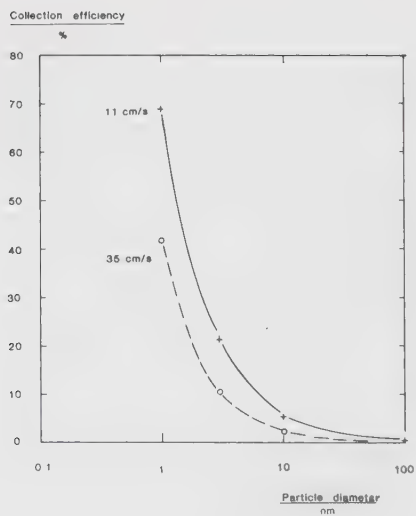


Figure 4.

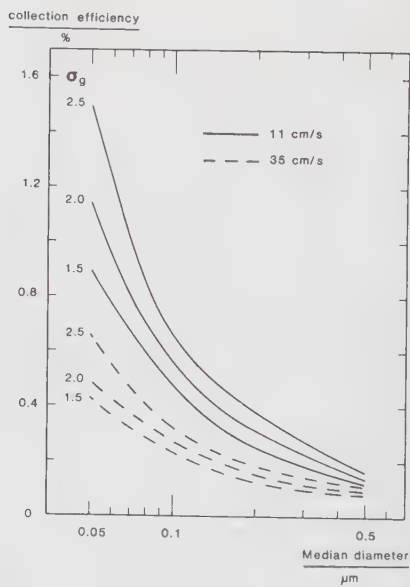


Figure 5.

Fraction of $\text{RaC}' - \alpha$ forwards

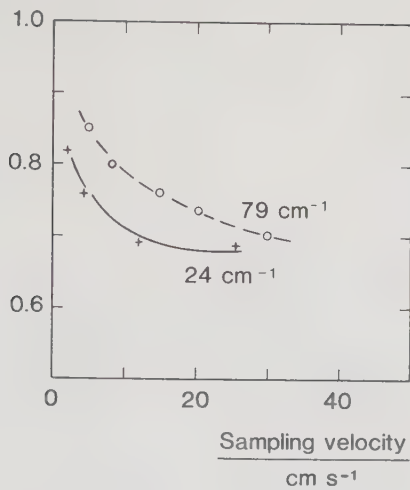


Figure 6.

Counts per channel

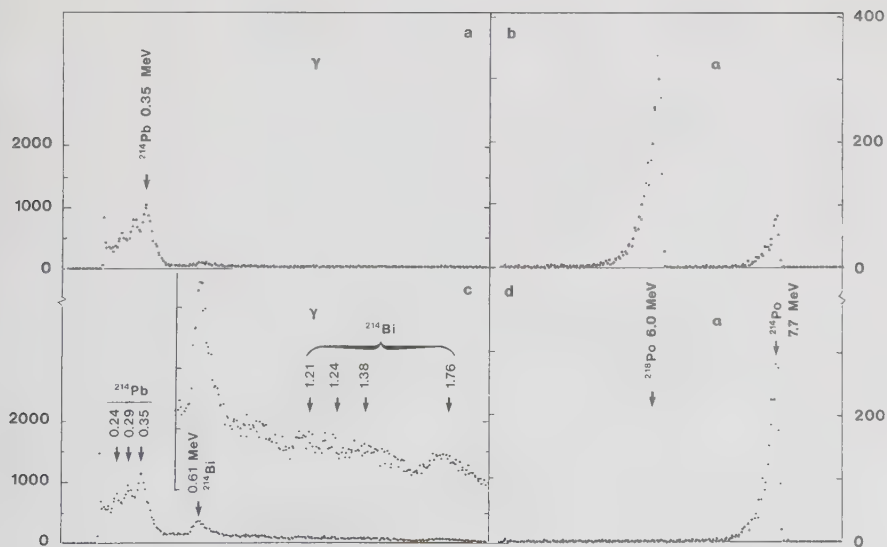


Figure 7.

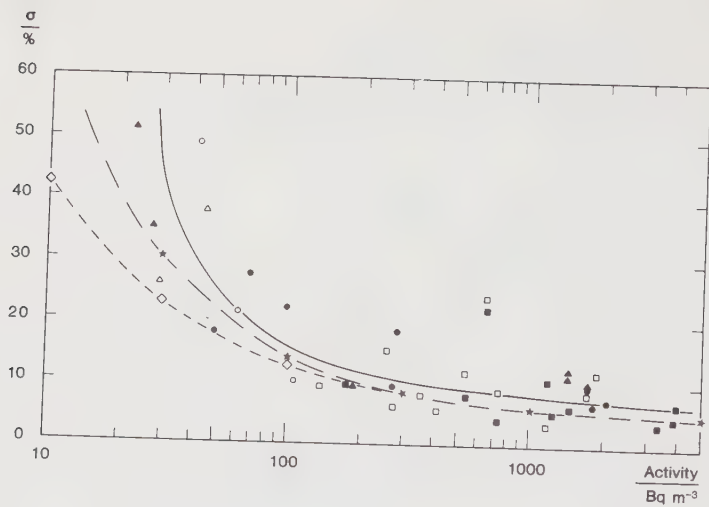


Figure 8.

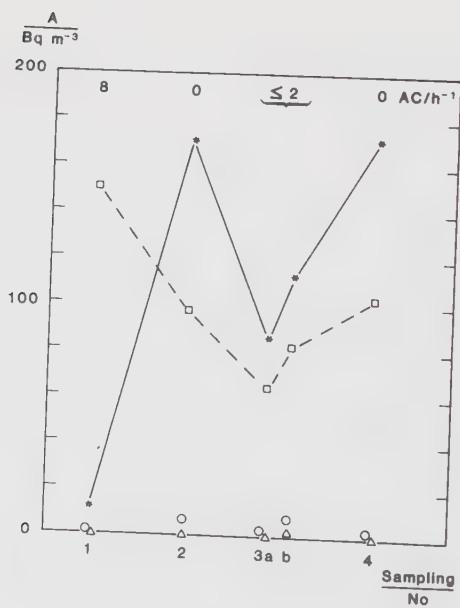


Figure 9.

Filter	-	-	-	G	G	G	-	-	G	C	W	-
AC/h ⁻¹	off	5	13	2	5	13	off	13	13	13	7.7	off
CNC/mm ⁻³	2.4	3.3	2	1	.27	.05	325	306	274	235	265	1.6
H _{JB} /μSv h ⁻¹	66	49	58	67	50	34	20	18	11	12	19	66
H _{JE} /μSv h ⁻¹	32	22	23	24	16	11	20	19	5	5.2	17	31
	a	b	c	d	e	f	g	h	i	j	k	l

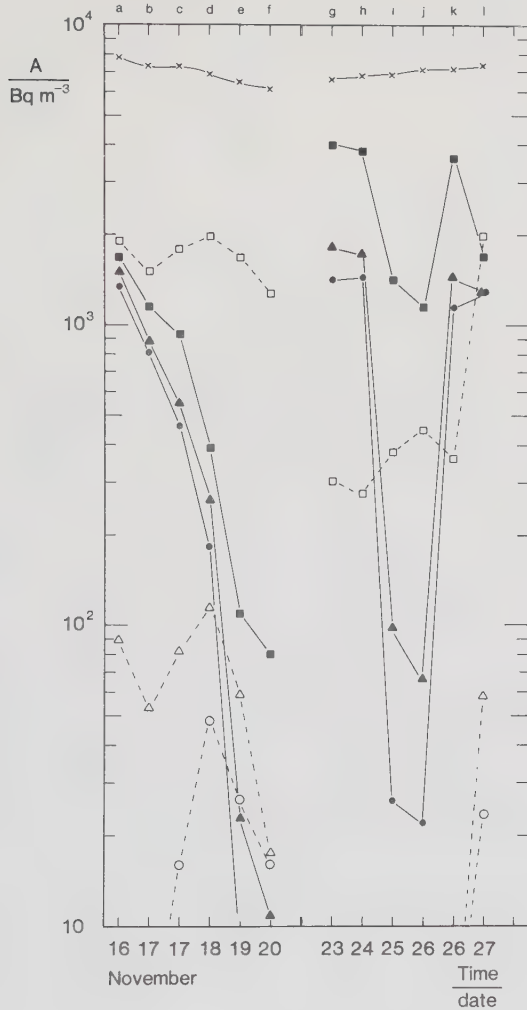


Figure 10.

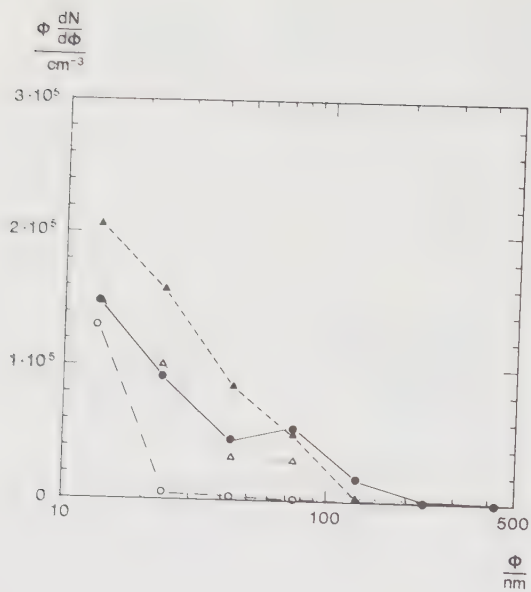


Figure 11.

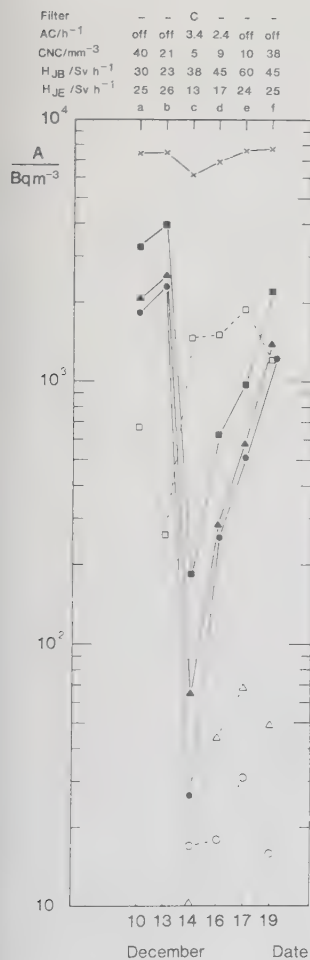


Figure 12.

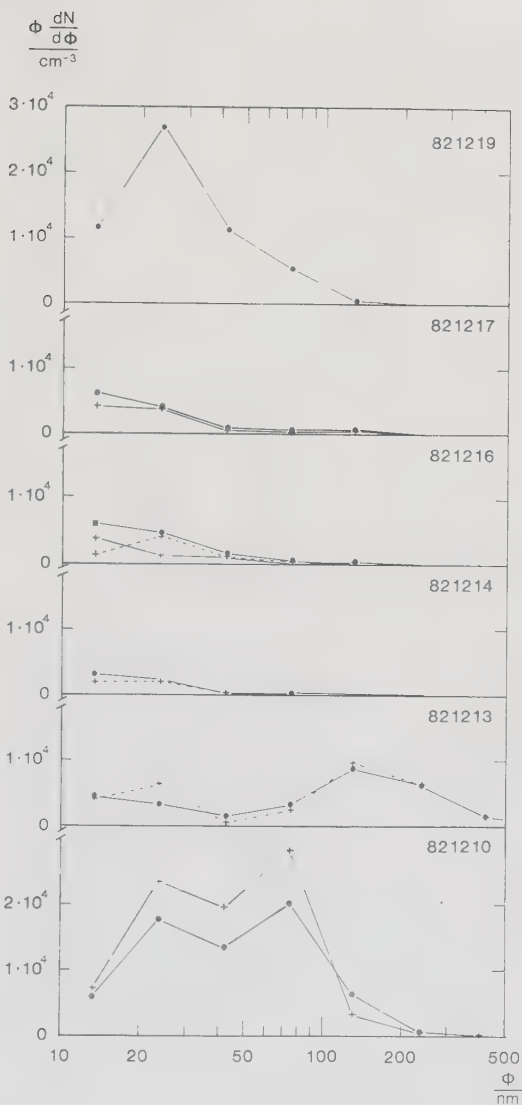


Figure 13.

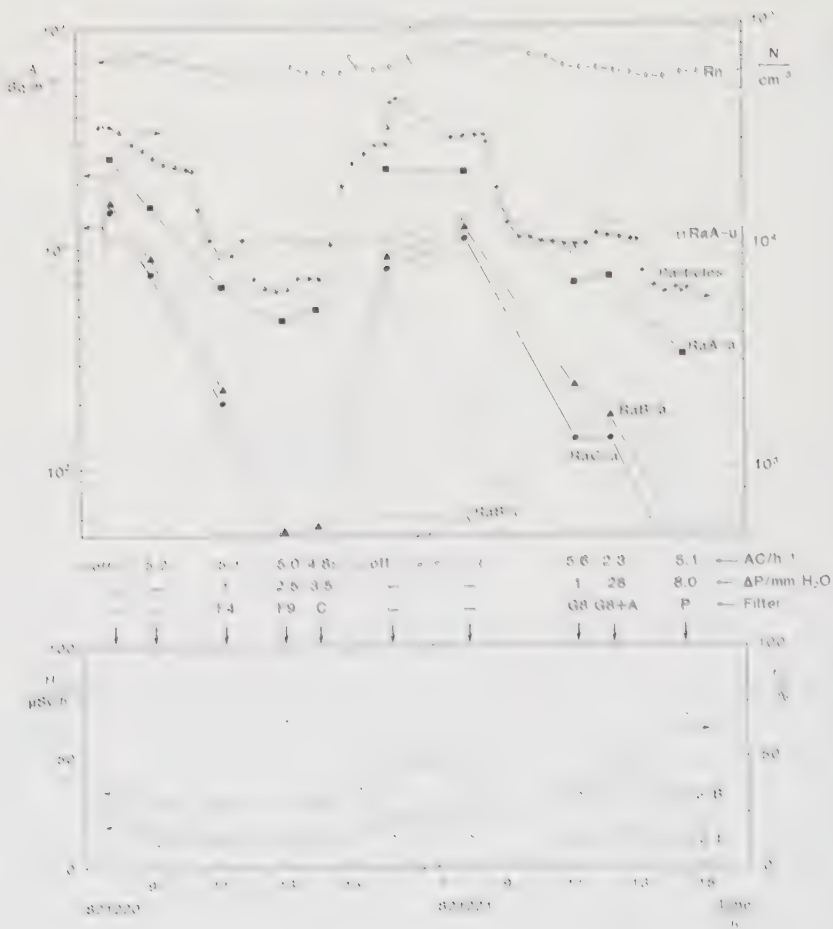


Figure 14.

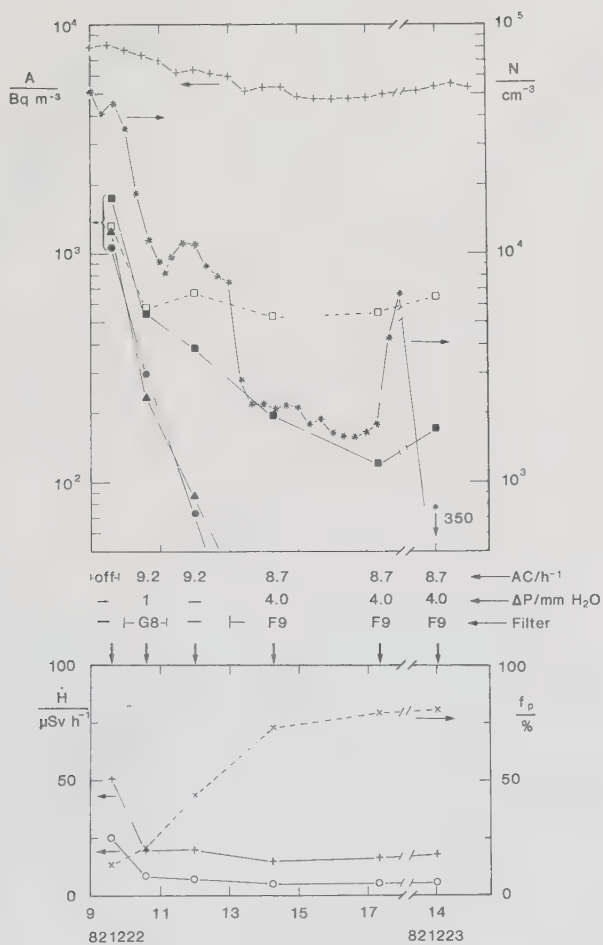


Figure 15.

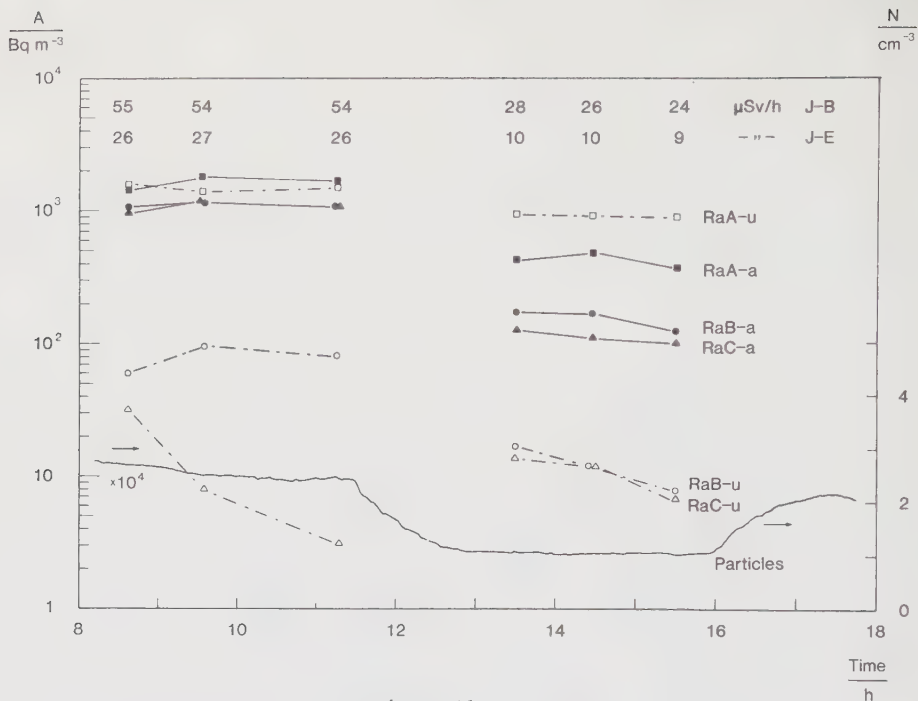


Figure 16.

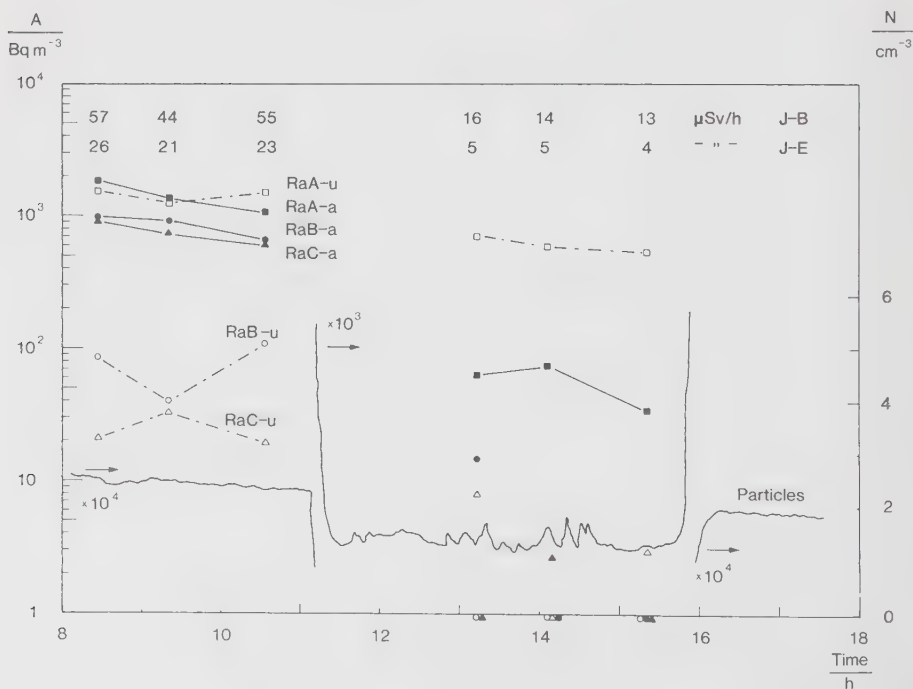
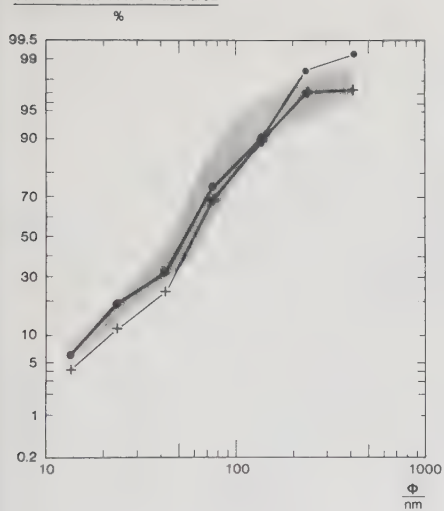


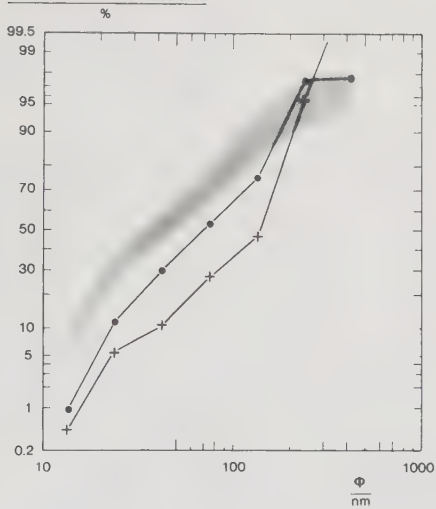
Figure 17.

Cumulative aerosol surface area



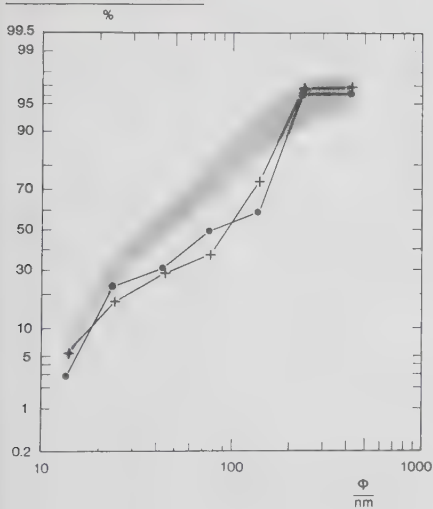
a.

Cumulative aerosol surface area



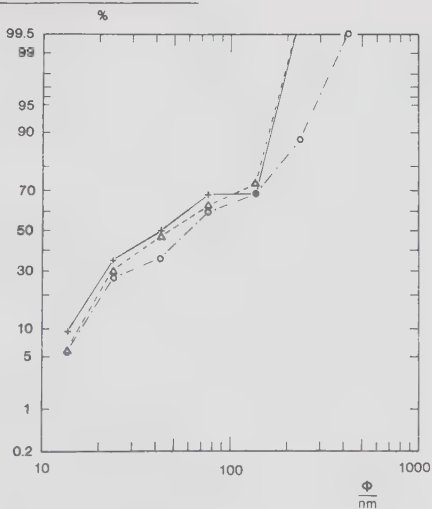
b.

Cumulative aerosol surface area



c.

Cumulative aerosol surface area



d.

Figure 18.

Relative effective dose equivalent

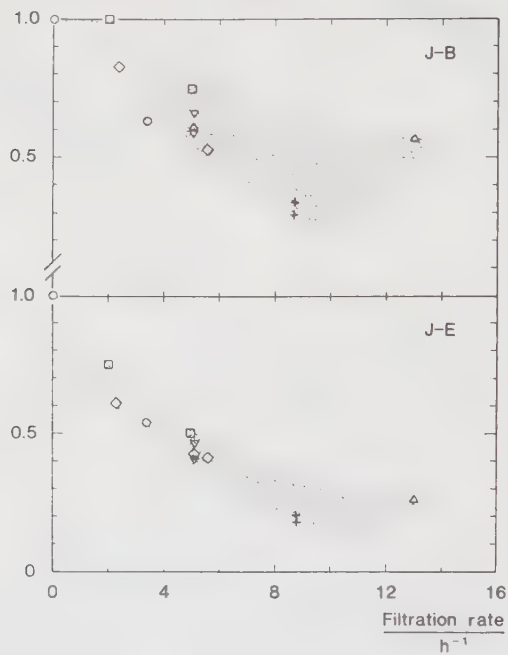


Figure 19.

^{222}Rn AND ^{210}Pb IN THE ARCTIC SUMMER AIR;
Assessment Techniques and Results from
the Swedish Expedition Ymer-80.

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1. INTRODUCTION.

^{222}Rn and its long-lived decay products have been proposed and used in several investigations as natural tracers in atmospheric processes. Review papers, e.g. Wilkening (1981), have been published covering this field. Several relevant articles are also found in the proceedings of the 'Natural Radiation Environment' conferences (NRE, 1968; NRE II, 1972; NRE III, 1980) and in the UNSCEAR report (UNSCEAR, 1982). ^{222}Rn and its progenies are members of the natural uranium decay series (Fig. 1).

The world-wide average exhalation rate of ^{222}Rn from soil, perma-ice regions excluded, is about $15 \text{ mBq m}^{-2} \text{ s}^{-1}$ (Wilkening & Watkins, 1976; UNSCEAR, 1982), and the corresponding figure from ocean water is $0.2 \text{ mBq m}^{-2} \text{ s}^{-1}$ (Jacobi, 1962). ($1 \text{ Bq} = 27 \text{ pCi}$).

The ^{222}Rn vertical concentration profile is to a great extent determined by the atmospheric stability. Typical ^{222}Rn concentration values in the surface air over land are only a few Bq m^{-3} , but much higher values can be found during temperature inversion (Nevissi & Schell, 1980). Gesell (1983) estimates the average concentration in surface air over the contiguous area of United States to 9 Bq m^{-3} . During normal turbulence conditions the radon concentration varies little with altitude within the planetary boundary layer of 0-2 km (Druilhet & Guedalia & Fontan 1980; Gesell, 1983) as theoretically predicted by Jacobi &

André (1963) and which is supported by measurements, e.g. those of Larson (1974).

The exhalation rate from land areas is reduced when the ground is frozen (Pearson & Jones, 1966). Several investigators have observed seasonal variation in the radon concentration in air, with a minimum in spring and summer (UNSCEAR, 1982), while Harley (1978) reported a maximum in mid-summer. The decreased air mixing during cold periods will increase the radon concentration in the lower air over land. This increase is counteracted by a decreased exhalation rate at low temperatures.

The exhalation rate of ^{222}Rn across the earth-air or a sea-air interface is mainly governed by the source strength - that is, the 'emanating' ^{226}Ra , and the effective diffusion coefficient. Several parameters, such as barometric pressure, soil porosity, soil humidity and wind speed/water turbulence will modify the exhalation of radon (Tanner, 1980; Roether & Kromer, 1978). As a mean, however, cf. figures given above, the exhalation rate from sea water is two orders of magnitude lower than that from soil. On land, sources of ^{222}Rn other than soil (for example vegetation, running water and coal burning) are not significant (Cohen, 1979; UNSCEAR, 1982).

The continents can be considered as large extended sources of ^{222}Rn . Continental air masses over the oceans can be traced by assessing the radon concentration in air (Larson, 1978; Wilkness & Rodgers & Swinnerton & Larson & Lamontage, 1979; Larson & Bressan, 1980).

At decay of ^{222}Rn , the short-lived decay products attach themselves to ambient aerosol particles. The details of this attachment process are quite complicated, and lead to somewhat conflicting experimental results (Ho & Hopke & Stukel, 1982). The half-life of attachment is about 1 minute or less in 'normal' continental outdoor air (Mohnen, 1969; Postendörfer & Mercer, 1980). This means that during normal conditions the majority of short-lived daughters are attached to aerosol particles. Only the first daughter ^{218}Po ($T_{1/2}=3.05$ minutes), can be expected to be significantly unattached, as has been experimentally reported by Gat & Assaf & Miko (1966).

With the possible exception of rainfall, most aerosol scavenging processes in the atmosphere take place slowly, compared to the physical half-lives of the short-lived daughters (Hicks, 1978; Finck & Persson, 1980). Shapiro & Kosowski & Jones (1978) measured large variations in the activity ratio $^{214}\text{Bi}/^{214}\text{Pb}$ in coastal air. The relative standard deviation in their mean ratio, 0.85, is about

30%, at a sampling height of 20 meter above the ground. The fairly large deviations from equilibrium obtained by Shapiro et al (1978) have been questioned by Larson (1979), who was unable to find any significant deviation from equilibrium in maritime radon daughter aerosols, including measurements under fog conditions.

The time scale relevant for long-range transport of air masses is days or weeks. In this time scale, it is obvious that the long-lived decay products of ^{222}Rn are of interest. It is justified to consider ^{222}Rn as a gaseous precursor to a ^{210}Pb -contaminated aerosol. The physical decay of ^{210}Pb ($T_{1/2} = 22 \text{ a}$) is of no significance compared to other scavenging processes, and therefore the air-activity concentration of ^{210}Pb is much lower than that of ^{222}Rn . A typical ^{210}Pb value in the lower atmosphere above the continents is 0.5 mBq m^{-3} (Jaworowski, 1969; Sanak & Lambert & Ardouin, 1980). Several authors have used measurements of ^{222}Rn (or its short-lived decay products) and/or the long-lived daughters to obtain valuable meteorological information (Moore & Poet & Martell, 1975; Mishra & Rangarajan & Eapen, 1980; Rahn & McCaffrey, 1979; Larson & Bressan, 1980). If the measurements are made far away from the radon source, i.e. the continents, the absolute value of the ^{222}Rn air concentration is a measure of the time passed since the air left the land. The ratio of the activity of ^{222}Rn to its long-lived daughters yields information about the magnitude of the aerosol scavenging processes experienced by the air mass in question.

During the organization of an expedition to the Arctic Sea, the authors' proposal to carry out ^{222}Rn and ^{210}Pb measurements in the air-chemistry programme was accepted. The ice-breaker, Ymer of Sweden (Figure 2), was to be used, and this Arctic venture was done to commemorate the famous Northeast Passage of the explorer Nordenskiöld and his ship, the Vega, 100 years earlier (Litell, 1980). Beside the air-chemistry programme, the authors' departments also participated in the water-research programme. The two legs of the expedition are shown in Figure 3.

Only a few measurements of ^{222}Rn and ^{210}Pb in Arctic air have been made. A typical summer value at Barrow, Alaska, is $100 \text{ } \mu\text{Bq m}^{-3}$ of ^{210}Pb (Rahn & McCaffrey, 1979). From Thule, Greenland, Patterson & Lockhart (1964) reported concentrations of ^{210}Pb during 1961-62 corresponding to a mean value of about $200 \text{ } \mu\text{Bq m}^{-3}$. More typical for maritime Arctic conditions are the concentrations on the islands of Amchitka, Alaska (54°N , 155°E). In summer, the air concentration of ^{210}Pb is about $2 \text{ } \mu\text{Bq m}^{-3}$ (Nevissi & Schell, 1980). From Russian measurements on Franz Josef land (cf Fig. 3), a range of $2 \text{ to } 75 \text{ } \mu\text{Bq m}^{-3}$ is given in the survey papers of Jaworowsky (1969). Rangarajan & Gopalakrishnan & Eapen (1976), who

indirectly cite the same Russian paper via its English translation, give a larger interval, 0.7-200 $\mu\text{Bq m}^{-3}$, and a mean value of 13 $\mu\text{Bq m}^{-3}$. To authors' knowledge, no measurements of ^{222}Rn in air during summer in Arctic maritime regions have been undertaken, but Lockhart (1962) measured ^{222}Rn at Kodiak Island ($57^{\circ}45' \text{ N}$, $152^{\circ}29' \text{ W}$) and Wales ($65^{\circ}37' \text{ N}$, $168^{\circ}03' \text{ W}$), Alaska. The relatively small land mass of Kodiak exhibits the lowest ^{222}Rn concentrations, a mean value in summer of about 200 mBq m^{-3} during 1953-1959. The corresponding figure at Wales was 400 mBq m^{-3} . Over the North Atlantic, the Norwegian Sea and Alaska, the same yearly mean radon-concentration values, 200 mBq m^{-3} , are estimated by the latest UNSCEAR report (UNSCEAR 1982).

2. EXPERIMENTAL SET UP.

Though the Department of Radiophysics has during several decades conducted outdoor gamma-measurements and biota-samplings in the Sub-Arctic environment, the Ymer expedition was a completely new challenge. Extremely low concentrations of activity were to be measured during rough ice-breaking. Dr. Lennart Granath and his collaborators at the Department of Meteorology, University of Stockholm, were responsible for the air chemistry programme and the design of the common air intake. The common air intake of the air-chemistry container consisted of a polythene pipe 15 cm in diameter. The inlet was situated about 23 metres above sea-level, and the pipe was bent 90 degrees in order to pass through the container horizontally (Fig. 4). To avoid contamination from the ship itself, the ship helicopters, etc., a sensitive condensation nuclei-counter monitored the inlet air and stopped the main sampling pump when a pre-set concentration level was exceeded. The air-flow through the main pipe was also stopped when the air-intake was situated on the leeward side of the ship's exhaust pipes. The two pumps sampling for ^{222}Rn and the radon daughters (short-lived and $^{210}\text{Pb}/^{210}\text{Po}$), respectively, worked continuously no matter what the status of the main sampling pump. All of the pumps were housed in a separate container, with a filtered air-exhaust.

2.1 ^{222}Rn experiments.

No radon-measuring equipment with a sensitivity suited to the low concentration levels anticipated in Ymer expedition was available for us. In the planning stage, two measuring principles were considered: high-volume sampling of short-lived daughters on filters (Larson, 1973), and trapping of radon on charcoal. The latter was chosen for several reasons: it was a direct radon measuring technique, easy to conform with the rest of the air chemistry programme, and appropriate commercial equipment was available (Radon concentrator RCTS-2, Johnston Labs., Cockeysville, Maryland, USA). It was, furthermore, possible to gain from earlier experience with indoor ZnS-vial methods and research by others in the field of charcoal adsorption (Bolmsjö & Persson, 1978). The drawback was that the method was not automatic. Only the modification of the concentrator will be presented here, since the use of such equipment has been adequately described by Lucas (1957) and also by Blanchard (1964). The charcoal in the radon-trap was changed to a type (Picatife G210, Pica, France) that had shown excellent qualities in earlier ^{155}Xe -investigations (Bolmsjö & Persson 1978). The Johnston Laboratories recommend a dry-ice solution to cool the charcoal-trap and the copper-coil water-trap. As carbon dioxide investigations of water and

air were a part of the scientific programme, dry ice was not allowed on board. Instead, dip-in freezing sonds (Julabo FT200, Juchheim Lab., Seelbach, W. Germany) were used to cool a solution of ethanole to about -40°C in thermally isolated flasks of the authors' own construction. Figure 5 shows the radon-concentrator installed in the laboratory container on board.

The trapped radon was transferred to ZnS-vials (LAC-II Johnston Lab., USA) in the usual manner by flushing the heated ($380-400^{\circ}\text{C}$) charcoal-trap with helium gas. The pulses from the ZnS-vials were counted for approximately 12 hours by PM-tubes connected to conventional pulse-counting electronics. To withstand the strain on board, the my-metal-shielded PM-tubes-housings were well secured and amply cushioned by plastic foam.

2.2 ^{222}Rn -decay products.

For the sampling of long-lived radon daughters, the inlet of an alpha-in-air monitor (Alpha-3, Eberline, Santa Fe, New Mexico, USA) was connected to the main air pipe. The air was continuously sampled at a rate of about $2.3\text{ m}^3\text{ h}^{-1}$ onto membrane filters (SM $5.0\mu\text{m}$, Sartorius, F.R.G.). A well-regulated pump (RAS-2, Eberline) drew the air through the filter with a constant flow-rate, irrespectively of the load of the filter. To enhance the counting efficiency, the filters were collimated to a diameter of 25 mm by a thin rubber ring. The filters were changed every 24 hours and stored for later laboratory ^{210}Pb -measurements. The monitor is equipped with a ruggedized surface-barrier detector, viewing the filter. It was thus possible to follow the short-lived daughter variations. The alpha-pulses, the majority of which originates from ^{214}Po , were fed into one channel of a 30-channel data-logger. (This connection was not effectuated until one month after the start of the expedition). The cycle-time of the data-logger was 30 seconds, and the alpha-counts received in each of these time-intervals were analyzed by a small computer (Granath, 1980). The minimum, maximum, median and lower quartal percentils, as well as the upper quartal percentil of the alpha-count rate during consecutive half-hour intervals were automatically calculated and printed out.

The filters brought to the laboratory were analyzed by alpha spectrometry for the long-lived daughters ^{210}Pb ($T_{1/2} = 22\text{ a}$) and ^{210}Po ($T_{1/2} = 138\text{ d}$). During the chemical procedure, which was a slight modification of that described in the HASL Procedures Manual (Harley, 1978), polonium deposited spontaneously onto nickel from an acid. As soon as was possible after the sampling, the membrane filters were wet-ashed and a Po-deposition performed. Before the filter was pro-

cessed, a known amount of ^{209}Po was added as a yield determinant: some losses inevitably occur, in particular during the wet-ashing procedure, since Po is a rather volatile element. The Po-deposited was measured by means of alpha spectrometry using a surface-barrier detector.

The solution, in which practically all the ^{210}Pb is left, was stored for about a year. ^{209}Po was then added once more and Po was again deposited and measured. The ^{210}Pb , as well as the ^{210}Po -content of the filter at the time of sampling, were thus obtained. Since the $^{210}\text{Po}/^{210}\text{Pb}$ -activity ratio in air is often of the order of 0.2, the delay between sampling and the first deposition should be short, preferably less than a week or two, in order to prevent ^{210}Po grown from ^{210}Pb to become a significant fraction of the total ^{210}Po -content, which would make the evaluation of the original ^{210}Po uncertain. Unfortunately, the delay mentioned was often too long, resulting in rather uncertain ^{210}Po -values.

3. EXPERIMENTAL ERRORS.

3.1 Evaluation and loss of ^{222}Rn -activity.

To calibrate the ZnS-vials and test the concentrator, radon gas from a ^{226}Ra -standard solution (Amersham, England) was used. The ^{226}Ra -bubbling system is similar to that designed by Rushing & Garcia & Clark (1964). By a re-cycling-dilution procedure, it was demonstrated that a few percent of the ^{222}Rn -activity could be lost in a concentration cycle, thus supporting the findings of Stehney & Norris & Lucas Jr & Johnston (1955) that small losses of radon occur in stop-cock-grease, anhydrous CaSO_4 and in water frozen in the water trap. The bolus of radon is moving forward in the air flow direction. A break-through of radon will thus take place, if large air-volumes are passed through the trap. Apart from the air-volume, several other parameters also influence the efficiency and break-through point of the charcoal: air-velocity, charcoal temperature and the amount of charcoal (20 grams in our trap). Known amounts of radon activity were diluted in air in large plastic bags to test the loss of radon in the concentrator as a function of the air-volume sampled. The results are given in Figure 6. The efficiency of radon-trapping on charcoal is strongly dependent on the volume of air through the trap. A charcoal temperature of -40°C and an air-velocity of about 1 litre/minute was used in the present investigation. The efficiency is plotted in Figure 6 as a function of air volume through the charcoal-trap. Up to about 1000 litres, the efficiency is almost 100 %, but will decrease to 50 % at about 4000 litres of air.

Charcoal and CaSO_4 contain ^{226}Ra , and will therefore release radon. It is important to know this background, especially when radon concentrations are low. The background release of radon from the collecting system has been measured in the laboratory to about 2 mBq per sample (23 hours collection), which is 10 % of the lowest value found during the expedition. All radon samples were corrected for the background of the system, and for the loss of radon at large air-volumes. The uncorrected mean ^{222}Rn -concentration, C_{Rn} , in air during the sampling time, t_s (minutes), was calculated from the accumulated number of net pulses, p , during the counting time, t_m (minutes):

$$\text{Eq. 1} \quad C_{\text{Rn}} = \frac{p \cdot t_s \cdot \exp(t_u/\tau)}{E \cdot V \cdot \tau^2 \{1 - \exp(-t_m/\tau)\} \{1 - \exp(t_s/\tau)\}} \quad (\text{Bq m}^{-3})$$

where

E is the counting efficiency (cpm Bq⁻¹),

V is the sampled volume (m³),

t_u is the time between sampling-stop and counting-start, i.e. the build-up time (minutes) and

τ is the mean life of ²²²Rn, 7942.2 minutes.

The assumption underlying Equation 1 is that the radon concentration is constant throughout the whole sampling-interval. The build-up time, t_u, has been kept at about 3 hours in order to achieve radon daughter equilibrium in the ZnS-vial. All vials were individually calibrated.

To evaluate the statistical error in the radon concentration, it is necessary to take into account that some of the counts are statistically dependent. The Poisson standard deviation, N^{1/2}, of a number of registered counts, N, has to be replaced by (kN)^{1/2} as an estimate of the actual standard deviation. Typical parameter values for the radon-222 measurements on board Ymer were t_m = 1000 minutes, t_u = 180 minutes, V = 2.5 m³ and E = 100 cpm/Bq, giving a value for k = 1.8. As pointed out by Key (1977), the simple calculation of k given by Sarmiento & Hammond & Broecker (1976) yields a value deviating only a few percent from the exact calculation outlined by Lucas and Woodward (1964), provided that the counting time is >300 minutes. The long sampling time (t_s = 22 h) then yields a statistical error less than 3% down to C_{Rn} = 0.01 Bq/m³, an error that is insignificant compared to other uncertainties.

The efficiency, E, used is rather low due to a high discriminator-level in the alpha-counter, in order to exclude electrical noise. A regular test of the background count-rate of the ZnS-vials on a daily basis was performed on board. The background correction, 0.07 cpm, was very small.

Repeated use of the ZnS-vials in the laboratory, taking radon-samples from a ²²⁶Ra-solution, gave a relative standard deviation of 5 % in the individual activity determination. (As in the case of most radon-samples on Ymer, the statistical counting error in this case was negligible). It is, however, not possible to apply this figure immediately to the Ymer measurements, since the

concentrator also has to be considered. On the other hand, the variability due to the transfer of radon from the standard solution to the ZnS-vial should be subtracted. A reasonable estimate of the non-systematic error (relative standard deviation) in the radon concentration values on board is therefore 5-10 %. An upper limit can be estimated from the extended period of low ^{222}Rn -activity concentration during July 5-19 (cf Figure 7, below). The mean value is 19.3 mBq m^{-3} , and the experimental standard deviation in the individual concentration values is 5.0. This latter figure includes the variation in radon source strength. The relative standard deviation in the ^{222}Rn -concentration levels should thus be considerably less than $5.0/19.3=0.26$.

The main source contributing to the systematic error in the radon concentration is the uncertainty in coal-trap efficiency at large air-volumes. Figure 6 shows an uncertainty of less than about ~15 % for V less than 3 m^3 . Assessing the concentration of ^{210}Pb and ^{210}Po in the air, the counting statistics are a dominating source of uncertainty. The shaded area given in Figure 13, below, for ^{210}Pb include an estimate of the systematic uncertainty in the analysis method, as well as the stochastic contributions, and correspond to one standard deviation.

3.2 Evaluation and loss of aerosol-activity.

To calibrate and test the surface barrier detector on board, a ^{239}Pu alpha source was utilized on a regular basis. The relative standard deviation, 8 %, in the data logger signal corresponding to the Pu-source, was acceptable, but much greater than predicted from plain counting statistics. The added fluctuation was caused by sensitivity drift and disturbances of the electronic equipment, extra background count-rates due to dirt on the detector, and non-deliberate changes in the counting geometry. Electronic disturbances from the ship itself made it impossible to evaluate the short-lived daughters on several occasions. Such disturbances were easily identified in the data-logger output, and were hence excluded from the calculations. To confine the evaluation to a situation of short-lived daughter equilibrium on the filter, the first three hours following a filter change were not considered. The activity of the short-lived daughters is presented below as " Bq/m^3 " but in this case the unit should be considered as only relative. The reason for this is that the calculation is based on the ^{239}Pu -alpha counting efficiency in a somewhat different geometry, and that radon daughter equilibrium is assumed. Since the objective of including the short-lived daughters in an on-line measurement was merely to achieve a better time-resolution, than could be obtained by the radon day-to-day sampling, the draw-

The method of long-lived daughter analysis has been described above. Before any activity results in absolute terms can be given, however, the eventual loss of activity in the sampling-filter and the air inlet-system must be considered. This loss is a function of the particle-size distribution as well as other characteristics of the aerosol, and the sampling geometry. Prior to the expedition, the collection efficiency of the Sartorius SM-filter was investigated. The same geometry and sampling rate on board were employed, and a Millipore AA membrane filter ($0.8 \mu\text{m}$) at low flow-rates served as a reference filter with a 100 % sampling efficiency. No significant activity loss in the Sartorius-filter was found. These results have later been confirmed by Liu & Pui & Rulow (1982), who investigated the sampling characteristics of several filter-media, among them $5.0 \mu\text{m}$ membrane filters, up to air flow velocities of 100 cm s^{-1} and over the particle-size range of $0.0035\text{--}1 \mu\text{m}$.

In order to calculate the activity losses in the sampling system, one must know the activity-size distribution and the details of the sampling geometry. In the planning stage of the expedition, it was assumed that aerosol activity losses ought not be a great problem, since the summer Arctic aerosol should exhibit the characteristics of a fairly well-aged aerosol, with large particles (> 1 micrometer) eliminated by sedimentation, precipitation, etc. and the very small particles (< 0.01 micrometer, the gas-to-particle conversion range) essentially eliminated by coagulation. The activity losses due to impaction, sedimentation and diffusion should therefore be insignificant. In the main sampling pipe the diffusional losses can be roughly estimated from the standard diffusion-tube equation (Gormley & Kennedy, 1949; Thomas, 1967). At the sampling-rate ($4 \text{ m}^3 \text{ min}^{-1}$) the percentual losses at the particle diameter of 0.001 micrometer are about 4 %.

The air sampling inlet geometry of the alpha-monitor Alpha-3 is rather complicated. Losses of activity in the sampling-head of the alpha-monitor have therefore been experimentally investigated in two ways. First, five open filters taken with a high-volume sampler on board have been compared to the alpha-monitor results. No detectable losses of ^{210}Pb could be seen in this material. Secondly, air filter samples alternately taken with the Alpha-3 and an open filter-holder, attached inside the ventilation-pipe to the tubing leading to the Alpha-3, were compared by alpha spectrometry. The aerosol in a 3 m^3 radon-room, connected in a closed loop with the large ventilation-pipe, was simultaneously followed by a condensation nucleus counter (CNC TSI-3030, TSI Inc., St. Paul, Minnesota, USA) and an electrical aerosol analyser (EAA TSI-3020, TSI Inc., USA). No significant losses of aerosol activity of either ^{218}Po , ^{214}Pb or ^{214}Bi

could be seen, comparing the Alpha-3 with the open holder. The result can be assumed to apply to the Ymer conditions, since the aerosol-size distribution in the radon-room is similar to the distributions reported from the expedition published by Jaenicke & Schütz (1982).

Two independent investigations of aerosol losses in the sampling system have been described above. No significant loss of activity is indicated. As a consequence, no loss-correction factor has been applied to the ^{210}Pb -samples from Ymer. The accuracy of the loss-tests is low, however, and ^{210}Pb losses of up to 10-15 % would not be revealed.

4. RESULTS AND DISCUSSION

In a simple model, with the continents acting as radon sources and the oceans neither as sources nor sinks of radon, the apparent age of the sea air can be calculated. This age is a measure of the time passed since the air-mass left the continent. The source 'strength' and the dispersion of the air-mass must be known, to perform an exact calculation of air age. From islands and peninsulas, the air-mass may, however, pick up so much radon, that the simple two-category model of oceans/continents is incorrect. A related phenomenon, also disturbing the "age" calculation, is the mixing of air with different origins in unknown portions. The dispersion of radon over the sea is difficult to assess, while scavenging by precipitation is insignificant, due to the inert properties of radon. The existence of many unknown parameters, e.g. the source-strength variation in time and space, make it reasonable to limit the discussion of radon levels over the sea to qualitative terms.

The same restricted attitude must also prevail when dealing with ratios of activity concentrations contra mean residence time calculations, as discussed by Martell & Moore (1974). In particular, the ratio of ^{210}Po to ^{210}Pb may yield erroneous aerosol mean residence times, since several different sources are responsible for the air-borne ^{210}Po , while atmospheric ^{222}Rn overwhelmingly dominates as a source of ^{210}Pb in air. One fundamental premise concerning residence time calculations, using quotients of activities from a decay series, is rarely mentioned in the literature: There must be a constant source-supply or a mother nuclide of residence time much longer than the daughter, if steady state formulas are to be used. Following a land air-mass out over the sea, the radon source is lost. When comparing e.g. ^{210}Pb and ^{222}Rn , the mean aerosol residence time for ^{210}Pb over the sea must therefore be much shorter than 5.5 days, the mean residence time for radon. In Section 4.2., below, the subject of residence time calculations is further elucidated.

In the following sections, the experimental results from the Ymer expedition are exemplified, followed by discussions on the strength of local sources, such as the sea and the ship, itself. Our radiological results are also compared to results from other parts of the air-chemistry programme, published by Lannefors & Heintzenberg & Hansson (1983).

4.1 Radon

The ^{222}Rn -concentrations determined are shown in Figure 7. The numbers in the figure refer to the air-mass trajectories presented in Figure 8. The trajectory data were obtained from the Kernforschungszentrum, Karlsruhe (Schütz & v. Holleufer-Kypfe, 1981). The twelve chosen examples show how, in most cases, a qualitative correlation can be seen between radon concentration and air trajectories: The radon concentration decreases with the time elapsed since the air-mass left land. It is clear from the examples in Figure 8, that, to a large extent, the sea-level trajectory alone can explain the radon concentration level: In the 48 hour perspective shown, it would seem that the influence from higher levels is negligible. Consider, e.g., trajectories Nos. 9 and 10. As soon as the surface trajectory originates from the sea, the radon concentration becomes low, although the 1.5 and 3 km altitude trajectories are still passing over Greenland.

During an extended period (trajectories Nos. 1-4) in July, the air-mass reaching the ship always came from the North Polar area. With a few exceptions, low wind-strengths were prevailing. The radon concentrations were remarkably constant, with a mean value of 21 mBq m^{-3} , and a relative standard deviation of only 0.24. From the end of July to the end of the expedition, the weather (and ship positions) is more variative and low "Arctic background"-levels, as well as typical continental air-levels are encountered. Some cases are not so clear-cut, so that we obviously need to know more about the pattern of "radon-sources" over large regions, and also the movements and dispersion of the air over periods longer than the 48 hours shown in Figure 8.

The average radon concentration value for the whole time of the Ymer expedition (all samples north of latitude 75°) is $75 \pm 21 \text{ mBq m}^{-3}$ (one standard deviation). The mean value for July is very low, $32 \pm 19 \text{ mBq m}^{-3}$.

In connection with the low radon values during July, it is of interest to investigate the Arctic Sea and its ice as a radon source. In Section 3.1, it was mentioned that the measuring system itself contributed with about 1 mBq m^{-3} (2 mBq per sample). To calculate the amount of radon emanating from the sea, one must know the concentrations of radon in the water and the sea-air gas exchange, and to translate the exhalation rate to an air concentration at different heights over the sea, considering the mixing processes (turbulent diffusion, etc.). The sea-air gas-exchange has been studied by Broecker & Peng (1974), and by Roether & Kromer (1978). The latter authors measured the concentration of radon in the sea ($58.6^{\circ}\text{N } 12.2^{\circ}\text{W}$) and used the radon deficiency in the surface-

water layers to assess the gas-transfer. Roether and Kromer derived a transfer-velocity of 10 cm h^{-1} at a wind-speed of 9 m s^{-1} , and indicated that the value is dependent on the square of the wind-speed. The latter was less than 13 m/s at the ship's position during July. The exhalation of radon should therefore be less than approximately $0.06 \text{ mBq m}^{-2} \text{ s}^{-1}$, if a typical radon concentration in water of 1 Bq m^{-3} is assumed.

In a simple one-dimensioned model, neglecting horizontal variation in exhalation rate, we have at steady state:

$$D \frac{\partial^2 C(z)}{\partial z^2} - \lambda \cdot C(z) = 0$$

where D is the turbulent diffusion coefficient, $C(z)$ the radon concentration at the height z above sea-level, and λ the effective elimination coefficient for radon. To calculate the radon concentration roughly, we made D constant and got:

$$C(z) = C(z = 0) \exp \{ -(z/D)^2 \cdot z \}$$

$$E = \int_0^{\infty} C(z) \cdot \lambda \cdot dz$$

E denotes the exhalation rate. The turbulent diffusion coefficient at, for example, 10-20 metres above the ground and the sea-level, varies over about three decades, depending on the atmospheric stability (Jacobi, 1962). During medium to strong turbulence, D can attain values in the range of $4\text{-}40 \text{ m}^2 \text{ s}^{-1}$. Draxler and Elliot (1977) gave a typical value of about $1\text{-}10 \text{ m}^2 \text{ s}^{-1}$ in the height interval of 10-100 metres over ground, during neutral stability. D -values in the range of $1\text{-}40 \text{ m}^2 \text{ s}^{-1}$, combined with the exhalation rate of $0.06 \text{ mBq m}^{-2} \text{ s}^{-1}$, support a near sea-level concentration in the interval of 10 to 2 mBq m^{-3} , if the above equations are used. More realistic calculations taking into account the height dependence of the turbulent diffusion coefficient, yield similar air concentrations. The usefulness of this rough calculation is limited, due to its large uncertainties. The choice of the exhalation rate is, of course, critical. The exhalation figure was estimated as an upper limit, valid for wind-speeds of about 13 m s^{-1} . When the sea is covered by ice, the exhalation rate will be lower. On the other hand, the ship will, during ice-breaking, disturb the sea, thereby increasing the gas-exchange. The conclusion from the crude calculations described above must be that a substantial fraction of the low radon concentrations encountered in July may originate from the sea itself. In

Sections 4.3 to 4.5 below, a few episodes covered by Figures 7 and 8 are discussed in more detail.

4.2 Radon Daughters

The monthly average concentrations of ^{210}Pb in air were 31 ± 15 , 89 ± 61 and 105 ± 57 $\mu\text{Bq m}^{-3}$ for July, August and September respectively, giving a grand average of 75 ± 28 $\mu\text{Bq m}^{-3}$. As in the case of radon, only ship-positions north of 75° have been used in the calculation of these mean values. Jaworowski (1969) estimated a yearly mean value for the Arctic region of 140 $\mu\text{Bq m}^{-3}$, based on measurements at Franz Josephland, USSR and Thule, Greenland.

Figures 9-11 show corresponding daughter- and radon values. The short-lived daughter values should be interpreted as being only relative, as explained in Section 3.2. The correlation coefficients from Figures 9-11 are: RnD-Rn : $+0.79$, $^{210}\text{Pb-Rn}$: $+0.55$ and $^{210}\text{Po-}^{210}\text{Pb}$: $+0.73$. As expected, the best correlation is seen between radon and short-lived daughters (RnD), and the least between ^{210}Pb and radon. In the latter case, the variation in ^{210}Pb -scavenging should be mainly responsible, since an overwhelming part of the atmospheric ^{210}Pb originates from ^{222}Rn in air.

As stressed by Moore & Poet & Martell (1975), the $^{210}\text{Pb}/^{222}\text{Rn}$ -ratio cannot be used to calculate the mean aerosol residence time close to the continental coasts, since the steady state condition is violated for air masses just moving from land to sea, or vice versa. For aged air over the oceans, however, a steady state model calculation is applicable, if the following premises are fulfilled (T_r = mean aerosol residence time):

- a. The exhalation rate of ^{222}Rn from the sea is negligible.
- b. The age of the air-mass (i.e. the time for transport from a land-mass) is much greater than T_r .
- c. T_r is much less than the mean residence time for radon (5.5 days).
- d. No mixing of air-masses with different ages has occurred.
- e. Measurements during and shortly (time $< T_r$) after precipitation are excluded.
- f. The only source of ^{210}Pb over the ocean is through decay of air-borne ^{222}Rn .

The mean aerosol residence time, T_r , over the ocean can then be obtained from

$$T_r = \lambda_r^{-1} = \lambda_{Pb}^{-1} \cdot (A_{Pb}/A_{Rn}) = 1.18 \cdot 10^4 \cdot (A_{Pb}/A_{Rn}) \quad (\text{days})$$

where λ_{Pb} is the physical decay constant of ^{210}Pb , and A denotes activity concentration. Deriving this formula for T_r , it is also assumed that $\lambda_{Pb} \ll \lambda_r$.

A typical value of A_{Pb}/A_{Rn} during the expedition has been 0.001. Inserting this value in the equation yields $T_r = 12$ days. As this result violates assumption c, on which the equation is based, the result can not be correct. In order to make a better estimate of the aerosol mean residence time, we have to look at the air mass over the ocean as a dynamic system. Making the following idealizing preassumptions:

- i. Steady state conditions prevail over land in an air-mass leaving land for the ocean at time zero.
- ii. The aerosol removal coefficient λ_r is constant in time and identical over land and sea.
- iii. The dispersion of radon and radon daughters over the sea is the same.
- iv. Radon over the sea is removed only by physical decay.
- v. Influences of the short-lived daughters, and the sea as a radon source are ignored.

the radon and the radon daughter activity-ratios can be calculated as a function of air-mass age over the sea. Figure 12 displays the activity ratios lead-to-radon and bismuth-to-lead, as a function of air-mass age over the sea. The right-hand scale shows the steady state mean residence time, assuming a long-lived mother nuclide.

It is obvious that no simple relationship between A_{Pb}/A_{Rn} and the mean aerosol residence time exists, even with the simplifying assumptions made. A ratio $A_{Pb}/A_{Rn} = 0.001$ points towards a value T_r in the interval of 4-7 days - a value much lower than the steady state value shown on the right axis in Figure 12. It is clear that the ratio $^{210}\text{Bi}/^{210}\text{Pb}$ is not either an estimator of T_r for $T_r > 2$ days, unless the air mass age is known.

4.3 Contamination from the ship.

As each of the five diesel engines of the ship consumes 500 litres of oil per hour, the exhaust gases constitute a contamination risk. The total exhaust gas

volume emitted per minute is about 500 m^3 . In spite of this, the radiological measurements on board are insensitive to contamination from the ship, as is shown in the following calculations.

Kulich (1977) measured the ^{226}Ra concentration in diesel oil. In seven oils of different origins he found values in the interval of $0.7\text{--}1.9 \text{ Bq m}^{-3}$. Supposing the Ymer oil had a ^{226}Ra -content of less than 2 Bq m^{-3} , a ^{222}Rn -content of less than $200 \mu\text{Bq m}^{-3}$ in the exhaust-gases results, if equilibrium is assumed. Even at the exhaust-gas outlet, the ^{222}Rn -concentration is thus much less than 15 mBq m^{-3} , the lowest value encountered on the Ymer.

Assuming radioactive equilibrium, and that 100 % (a clear overestimation for ^{210}Pb) of the activity was emitted with the exhaust gases, the concentration of ^{210}Pb and ^{210}Po would also be less than $200 \mu\text{Bq m}^{-3}$. The ratio to the lowest ^{210}Pb -concentration met is approximately 10, but the dispersion of the exhaust-gases, before reaching the aerosol air intake, guarantee that contamination with ^{210}Pb or ^{210}Po from the diesel engines can be no problem.

Another, and independent, way of estimating the significance of the ^{210}Pb contamination risk is to use the stable lead emission rate of the Ymer engines, together with a reasonable ^{210}Pb -to-stable-lead-ratio. (It has not been possible to find any published values for the ^{210}Pb - or ^{210}Po -contents of diesel oils). Lannefors and collaborators analysed several filter samples from the exhaust-pipes of the Ymer, and got an emission rate of stable lead of less than $0.6 \mu\text{g m}^{-3}$ from one engine (Lannefors & Heintzenberg & Hansson, 1983). The value includes both coarse and fine particles. Ratios of ^{210}Pb -to-stable-lead in lichens have been measured earlier by two of the authors (Persson & Holm & Lidén, 1974). Old lichen samples (taken during the 1800's in Sweden) show a typical ratio of 70 Bq mg^{-1} radiolead-to-lead. The lichens are exposed to the atmospheric ^{222}Rn and ^{210}Pb , giving high ^{210}Pb /lead ratios. Other examples of ^{210}Pb /lead ratios are $<5 \text{ Bq mg}^{-1}$ in soil (Kodiara et al, 1980), $1.0 \pm 0.6 \text{ Bq mg}^{-1}$ in glacier ice from 1920-1940, and 0.001 Bq mg^{-1} in metallic lead (Jaworowski, 1969). A 65 year old lead plate had only 60 Bq mg^{-1} (Jaworowski, 1969). It is reasonable to assume, due to the absence of contact with the atmosphere, that the radiolead/lead-ratio in oil is substantially smaller than 1 Bq mg^{-1} . The exhaust-gas concentration of ^{210}Pb should consequently be considerably less than 3 mBq m^{-3} from all of the five engines.

The above arguments are confirmed by the experimental results from the Ymer. Lannefors & Heintzenberg & Hansson (1983) identified two of their samples as 'ship contaminated'. This classification is based on characteristics of the

sulphur and soot size distributions in comparison with the filter samples from the exhaust-pipes of the ship engines. Radiological and stable element results covering the two ship-contamination samples (P4 and P23) are shown in Figure 13. On the first occasion, around the 6th of July, there was no increase in radon or lead-polonium whatsoever. During the second contamination the radon sampling was only partly in effect, but again there was no significant increase in the radioactive lead- or polonium levels.

These results justify the decision taken before the expedition, that the radiological samplings should be performed continuously, regardless of wind direction.

4.4 Impact of sea spray.

On a few occasions, mainly when navigating in open sea, sea-salt deposited on the barrier-detector surface, thus increasing the background and excluding some alpha particles from reaching the detector. After cleaning the ruggidized detector-surface, the sensitivity and background were again normal.

Two samples (P22 and P33) exhibited the characteristics of sea-spray (Lannefors & Heintzenberg & Hansson, 1983). The first sample was taken in open sea at an average wind-speed of 12 m s^{-1} and sample P33 was obtained in pack ice at about 6 m s^{-1} . On both of these occasions, the short-lived daughter signal was disturbed by sea-salt on the detector (cf Figure 13). Since the air-flow through the filter was kept constant by the air-flow regulator, the sampling of the long-lived lead and polonium activities continued normally.

In the sample P22, the concentration of chlorine is $8 \mu\text{g m}^{-3}$. No peaks in the ^{210}Pb - or ^{210}Po concentrations are noticeable, an experimental fact supported by the following crude calculation. Assuming a chlorine concentration in air of about $10 \mu\text{g m}^{-3}$, the concentration ratio of sea-to-air is $5 \cdot 10^{-10}$, if the bulk sea-chlorine concentration is $\sim 20 \text{ kg m}^{-3}$. The ^{210}Pb concentration in the water-samples from the Ymer is 0.3 to 1.6 Bq m^{-3} , close to the range of 1.7 - 2.0 for ocean surface waters published by Rama & Goldberg (1961). About 96% of the ^{210}Pb in the ocean water is dissolved (Bacon & Spencer & Brewer, 1980) as chlorine is, and it is reasonable to apply identical concentration ratios (air-to-sea concentrations) for both chlorine and ^{210}Pb . Sample P22, yielding the highest chlorine concentration during the Arctic cruise, then corresponds to the ^{210}Pb air concentration of about 0.5 nBq m^{-3} , a level totally negligible, compared to the

lowest Ymer values encountered. Two conclusions can be drawn from this circumstance. Firstly, the air sampling of ^{210}Pb on board the Ymer was in actual practice completely insensitive to contamination from the sea, and secondly, the source of the ^{210}Pb in Arctic background samples was not the sea.

4.5 Long-range transport.

Two long-range transport episodes have been identified by Lannefors & Heintzenberg & Hansson (1983) in samples P14 and P24 (Figure 13). The first one lasted only a few hours during the night between August 4 and 5. The development of the episode is best illustrated by the short-lived daughters, due to the better time resolution. Trajectory No. 6 in Figure 8 reveals that the surface air, reaching the ship, passed over Scandinavia less than two days earlier. This 'young' history of the air thus fully explains the high ^{222}Rn - and short-lived daughter levels. The episode (our sample No.41, cf. Fig. 13) was, however, not accompanied by a ^{210}Pb -peak. The explanation for this is most likely the precipitation period between the 4th of August at 12:00 GMT and the 5th of August at 00:00. A typical scavenging rate constant in moderate rain is about 1 h^{-1} , hence the precipitation period mentioned can eliminate a major fraction of the ^{210}Pb -activity.

The second incidence of long-range transport occurred on August 15-16 and elevated concentrations are seen in all values except for chlorine, cf Figure 11. Compared to the episode during August 4-5, the radon peak is lower, indicating somewhat older or more dispersed air. The distinct peak in ^{210}Pb and ^{210}Po , and the high Po/Pb - and Pb/Rn -ratios, bear the signs of an air-mass exposed to low scavenging rates. The air origin is the Atlantic Ocean (Fig. 8, trajectory No. 7), and the weather maps indicate dry weather in the period preceding the episode.

The long-range transport during August 4-5 is shown in more detail in Figure 14. The minimum, median and maximum of the activity concentration (cf Sections 2.2 and 3.2) of short-lived radon daughters, and the CN-values taken from the data-logger print-out, are displayed on a logarithmic scale. The light scattering coefficient, σ_{sp} , a measure of the mass of airborne materials, is included as the recorder output in the lower part of Figure 14. The extreme maximum and minimum values of the radon daughters immediately after 8.30 are due to the filter-change. (The high values are caused by light falling on the surface barrier detector). Two CNC-peaks of very short duration are found just before the main episode. The origin of these peaks ought to be a 'young' source,

presumably the ship itself, giving small particles of little total mass and low radioactivity content. The episode of long-range transport starts at about 19.00 GMT, with rapidly increasing CNC, σ_{sp} and RnD. A good time resolution of the CN-detector is given by the recorder output. The episode is apparently connected to a fast change in wind-direction, as displayed in the upper part of Figure 14, and during the change there is a short duration of wind from the south (180 degrees). The correlation between the light scattering coefficient, σ_{sp} , and the radon daughter activity concentration (RnD), is highly positive. The median values have the following correlation coefficients: $r_{RnD-CN} = 0.13$; $r_{RnD-\sigma_{sp}} = 0.89$ (from the 4th of Aug., 18.00 GMT, to the 5th of Aug., 3.00 GMT).

Another episode is shown in detail in Figure 15. On this occasion, a Danish military base, situated in Northern Greenland, contaminated the air around the ship and caused a shut-off of the sampling through the main pipe. The high CNC-values at 3.00-9.00 GMT originated from the oil-heated plant on the base, and in the time interval between 8.30-9.00 GMT, the smoke reached the ship. (Unfortunately, this time coincides with a filter-change in the radon daughter detector). The broad peak in CN-concentration during 3.00-8.00 GMT has no correspondence in radon daughter values. The reason for this has already been discussed in Section 4.3; (the low radioactivity content of oils). The conclusion is therefore that local settlements in Arctic region constitute no radioactive contamination risk. The only exception from this rule should be a coal-consuming settlement. The correlation between the CN-, σ_{sp} - and RnD-values in Figure 15 is very small. A comparison of the median values yields the following correlation coefficients: $r_{RnD-CN} = -0.22$; $r_{RnD-\sigma_{sp}} = 0.03$. The fog banks, designated by an 'f' in Figure 15, affect the CN-counter, but not the surface barrier detector. The passing banks of fog caused drastic drops in the CN-value.

5. CONCLUSIONS

Radon sampling on a charcoal trap is a direct method with high sensitivity and reliability, albeit manpower-demanding if frequent sampling is desired. Experience of the ^{210}Pb - and ^{210}Po -sampling shows that volumes larger than 50 m^3 should be analysed, and that a rapid (less than a few weeks) separation for the first ^{210}Po -assessment must be done, for optimal accuracy in the results. All electronics, PM-tubes, counters, etc., have withstood the rough environment on a large icebreaker navigating in heavy ice, though the on-line alpha-system was occasionally subject to disturbances.

A good qualitative agreement (an inverse relationship) between radon-levels and the time since the air left larger land areas was found. In the short time-perspective (48 hour trajectories), the surface trajectories alone seemed to affect the radon concentration values. During an extended period during July, the air concentration of radon was persistently low, with a mean value of about 20 mBq m^{-3} . Significant contributions from sea-emanating radon during this Arctic background-period cannot be excluded. The mean radon concentration for the whole expedition (July 7 - September 23, 1980) was $75 \pm 21\text{ mBq m}^{-3}$ ($\pm 1\text{ s.d.}$) and the average ^{210}Pb -concentration was lower by a factor of 1000, $75 \pm 28\text{ }\mu\text{Bq m}^{-3}$. In these mean values, only samples taken north of latitude 75°N are included.

Steady-state models are unsuitable for mean aerosol residence-time calculations in ocean air. Time-dependent calculations indicate a mean aerosol residence-time of 4 to 7 days in Arctic air.

The semi-continuous sampling of short-lived daughters has proved successful, giving a better time-resolution than the 24-hour-radon-sampling, though somewhat sensitive to electronic disturbances and sea-salt.

The comparison with results for stable constituents, like sulphur and chlorine, reveals that both ^{222}Rn and its long-lived daughters were insignificantly contaminated by the ship's exhaust-emission, emission from local settlements or radioactivity of the sea-salt particles. The comparison also shows that large sources of ^{222}Rn do not necessarily emit anthropogenic sulphur. During the Arctic summer, Greenland seems to be such an example.

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Figure captions.

Fig. 1. The natural uranium decay series.

Fig. 2. The ship Ymer.

Fig. 3. A map of the expedition Leg 1 and Leg 2.

Fig. 4a. The air chemistry laboratory container, second from the right, with the smaller pump container on its side. The black vertical pipe is the air inlet of the air chemistry programme.

4b. A close up of the air inlet.

Fig. 5. The radon concentrator on board. The air inlet tubing is seen on the very left connected to valve no. 1. The two cooling flasks in the lower part of the picture are adapted to the copper coil water trap (no. 1) at the inlet and to the charcoal radon trap (no. 3). (The photo is taken before the extra valve between trap no. 1 and trap no. 2 was installed).

Fig. 6. The trapping efficiency of the charcoal trap as used on board, as a function of air volume through the trap. Test model 1 implies a repetitive concentration and dilution technique, involving the same sample, while model 2 makes use of continuous sampling from a large radon room. The charcoal temperature is between -35°C and -40°C .

● Test model 1. $0.5-0.7 \text{ dm}^3 \text{ min}^{-1}$

□ ---- " ---- $1.3-1.5$ -- " --

x Test model 2. $0.7-0.9$ -- " --

△ ---- " ---- $1.3-1.4$ -- " --

Fig. 7. Radon concentration versus time. The number in the circles correspond to air trajectories in Figure 8. ($100 \text{ mBq m}^{-3} = 2.7 \text{ pCi m}^{-3}$).

- Fig. 8. Examples of air trajectories during the expedition as calculated by Schütz & v. Holleufer-Kypfe (1981). All maps are numbered to ease the comparison with Figure 7.
- Fig. 9. Short-lived radon daughter concentrations (RnD) versus concentration of ^{222}Rn . Only samples north 75°N are included. The correlation coefficient is + 0.79.
- Fig. 10. Concentration of ^{210}Pb plotted against ^{222}Rn ditto, for all samples north 75°N . The correlation coefficient is + 0.55, and the mean ratio $^{222}\text{Rn}/^{210}\text{Pb}$ is 1000.
- Fig. 11. Corresponding concentrations of ^{210}Pb and ^{210}Po in all air samples taken north 75°N . The mean ratio $^{210}\text{Po}/^{210}\text{Pb}$ is 0.2, below zero values excluded. The correlation coefficient including all points is + 0.73.
- Fig. 12. The time development of the activity ratios in air, for different mean aerosol residence times, $T_r = \lambda_r^{-1}$. The radon source is zeroed at time $T = 0$. T_s is the aerosol residence time estimated from the activity ratio, assuming a steady state model with a long-lived mother nuclide. A = activity. Indices Pb, Bi and Rn refer to ^{210}Pb , ^{210}Bi and ^{222}Rn respectively.
- Fig. 13. Examples of results from the Ymer air chemistry programme. The upper half of the figure displays non-radiological concentrations as adopted from Lannefors et al (1983), and the lower half shows the corresponding radiological values. The uncertainties (shadowed areas) and different episodes covered by the Figure, are discussed in the text. The non-marine component of sulphur is marked with a broken line. (Index f = fine mode, diameters 0.05-2 μm ; index c = coarse mode, diameters > 2 μm).

Fig. 14. Minimum-, median- and maximum values of the short-lived radon daughter concentration (RnD) and the condensation nuclei concentration (CNC) during an episode of long-range transport. The recorder signal (r.s.) is shown in the lower part of the Figure. 90 and 180 degrees wind direction denote winds coming from the east and south, respectively.

fc = Filter change in the radon daughter detector.

----- = CNC recorder, 1 volt corresponds to 100 particles per cm^3 .

——— = σ_{sp} recorder signal, light scattering coefficient at 525 nm (the small nephelometer). 1 volt is about $5 \cdot 10^{-5} \text{ m}^{-1}$.

..... = RnD recorder signal.

Fig. 15. The half hour median values of CNC, RnD and σ_{sp} (at 550 nm), covering the contamination period from Station Nord, Greenland. The sampling through the main pipe is off between 4.50 and 10.10 GMT.

f = fog banks passing the ship.

fc = filter change in the radon daughter detector.

(The CNC and σ_{sp} values in the figure have already been published by Lannefors & Heintzenberg & Hansson 1983).

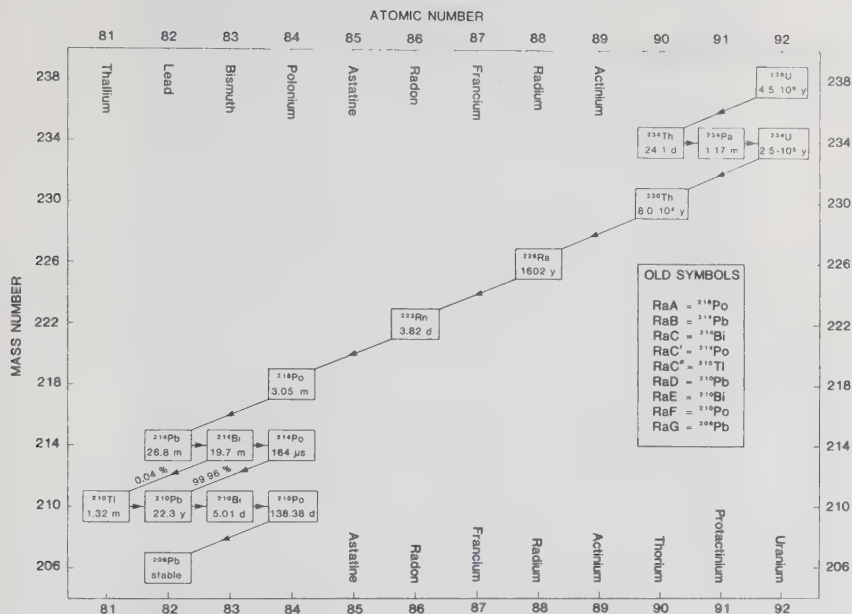


Figure 1.



Figure 2.



Figure 3.

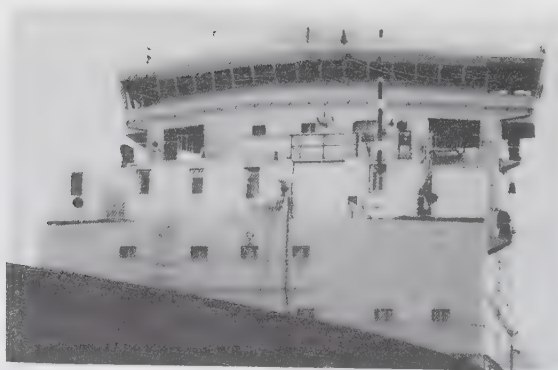


Figure 4a.



Figure 4b.

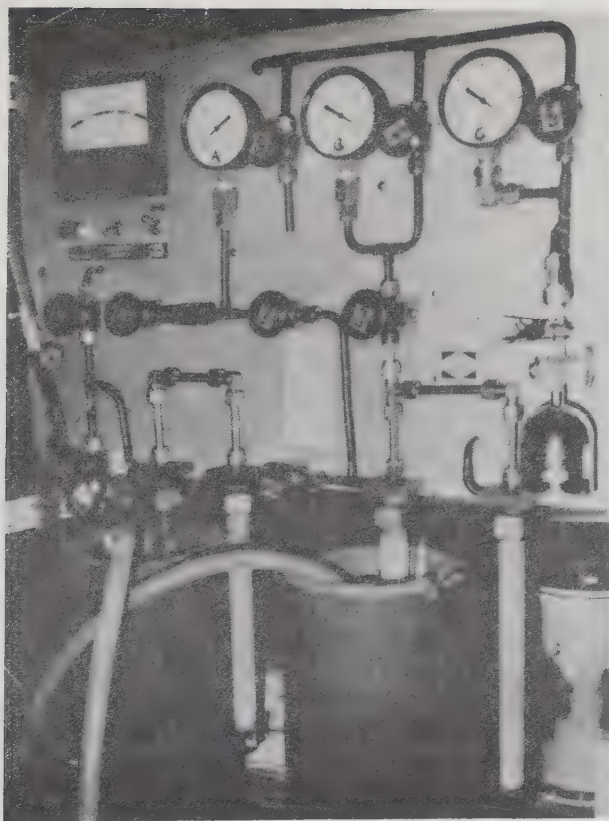


Figure 5.

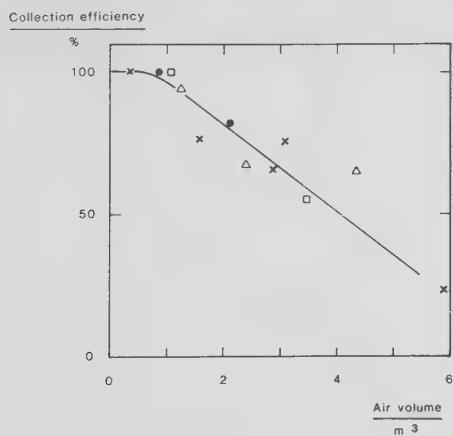


Figure 6.

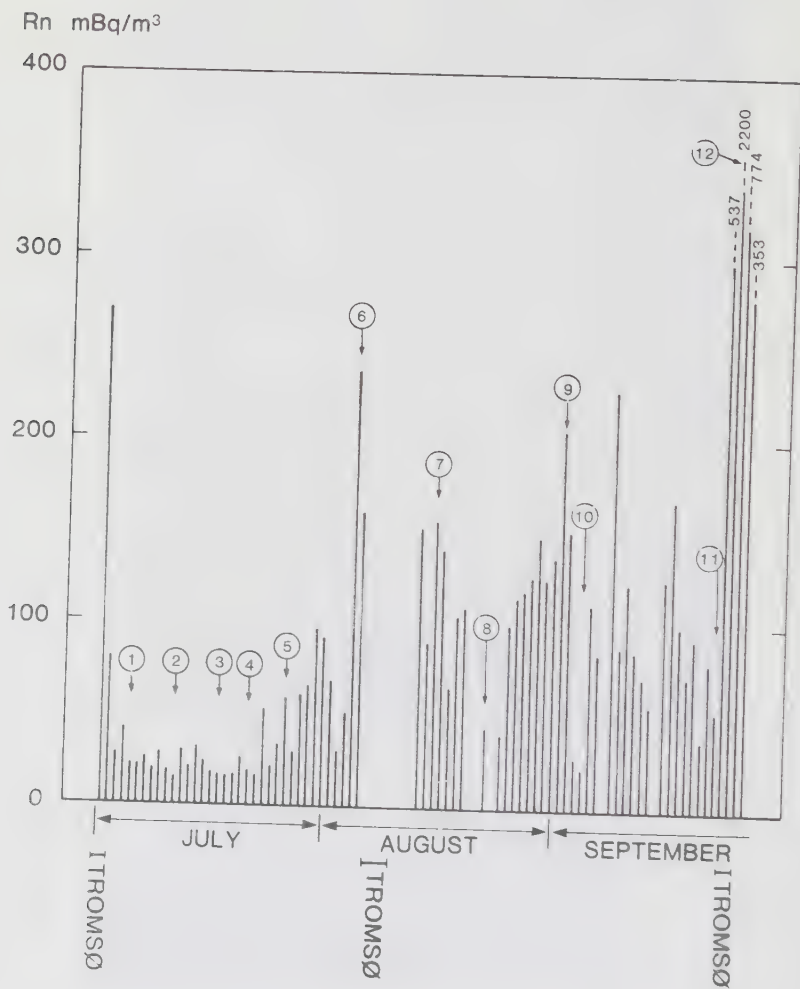


Figure 7.

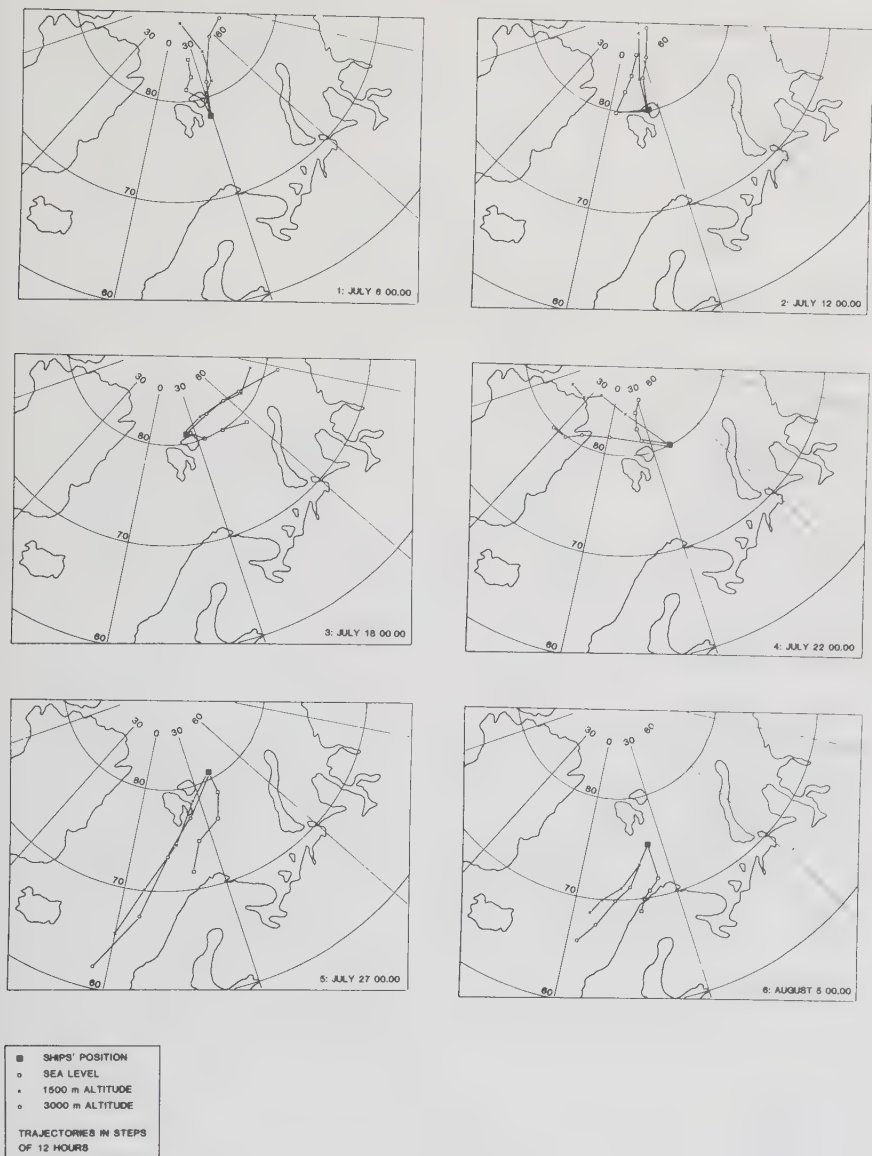


Figure 8.



Figure 8 continued.

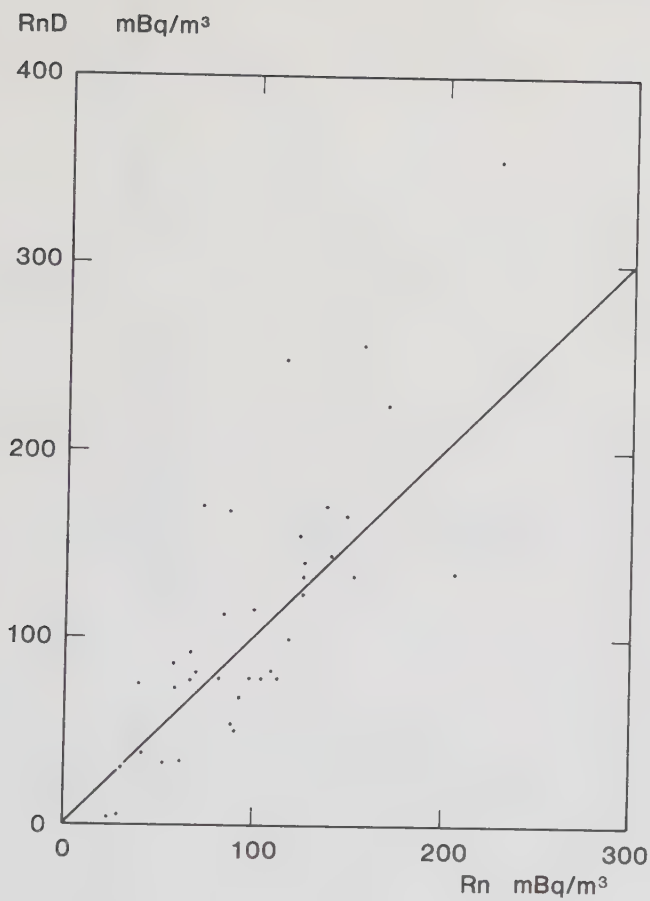


Figure 9.

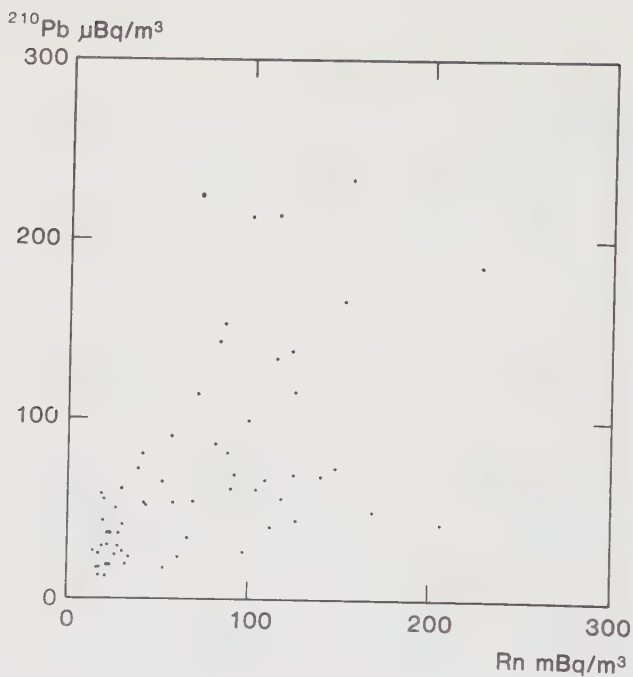


Figure 10.

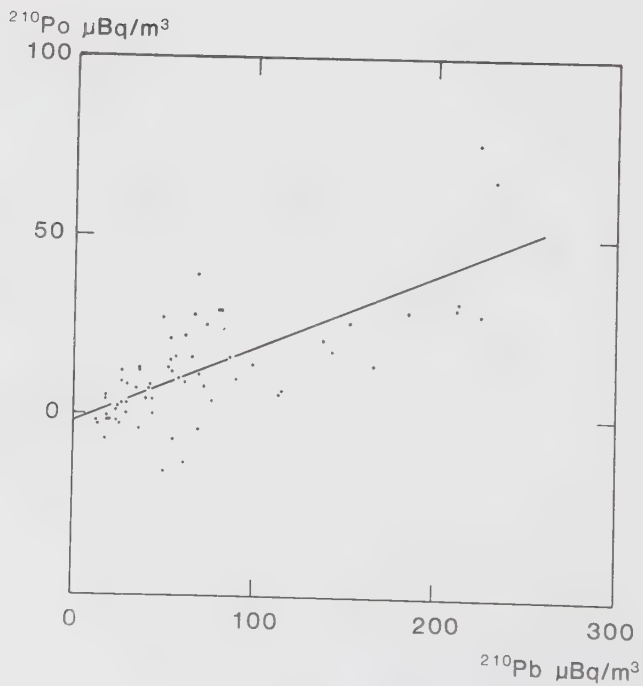


Figure 11.

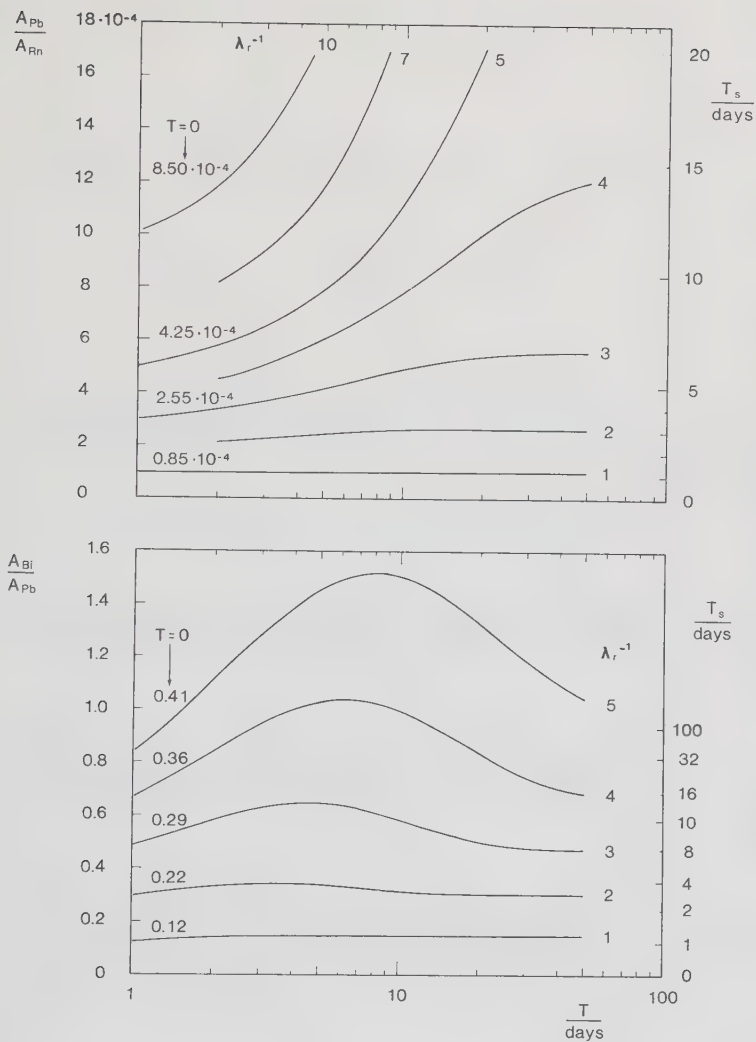


Figure 12.

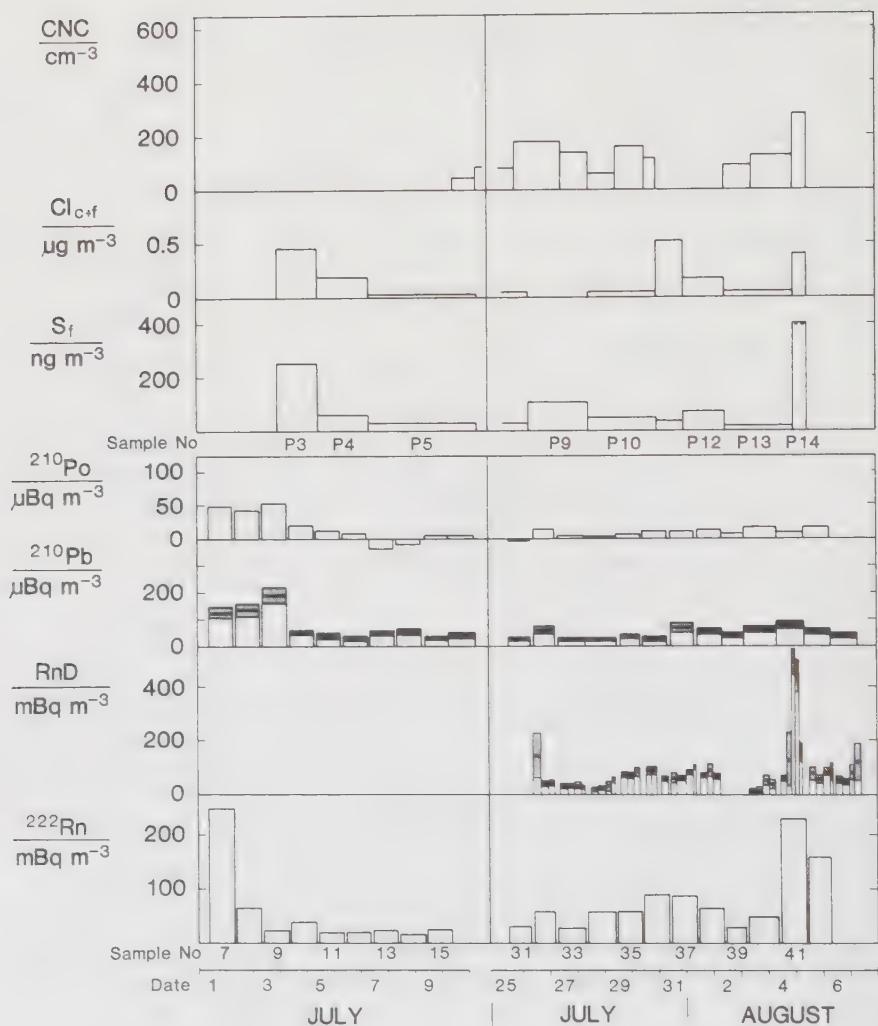


Figure 13.

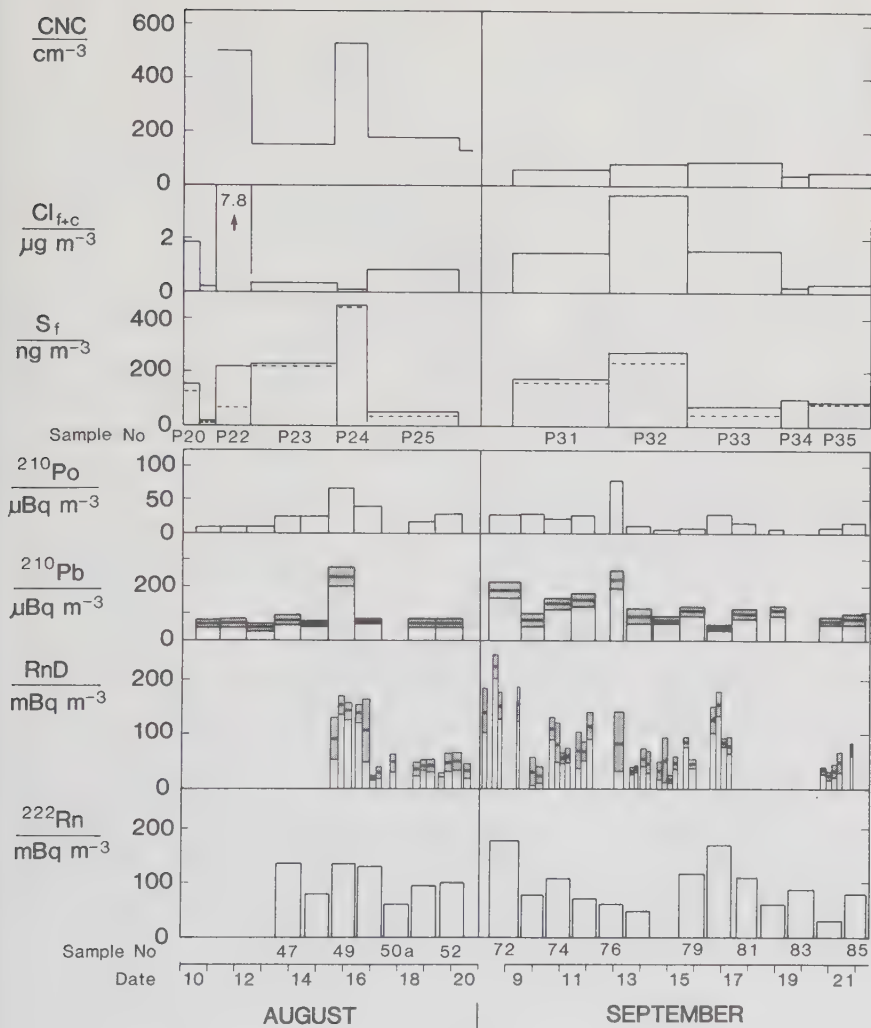


Figure 13 continued.

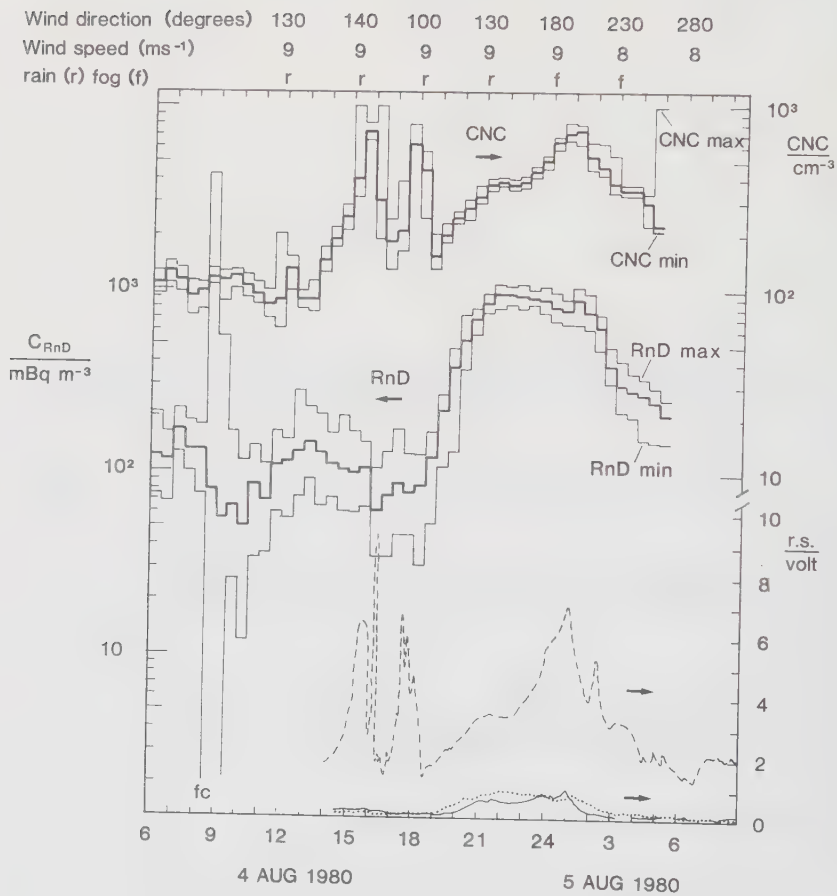


Figure 14.

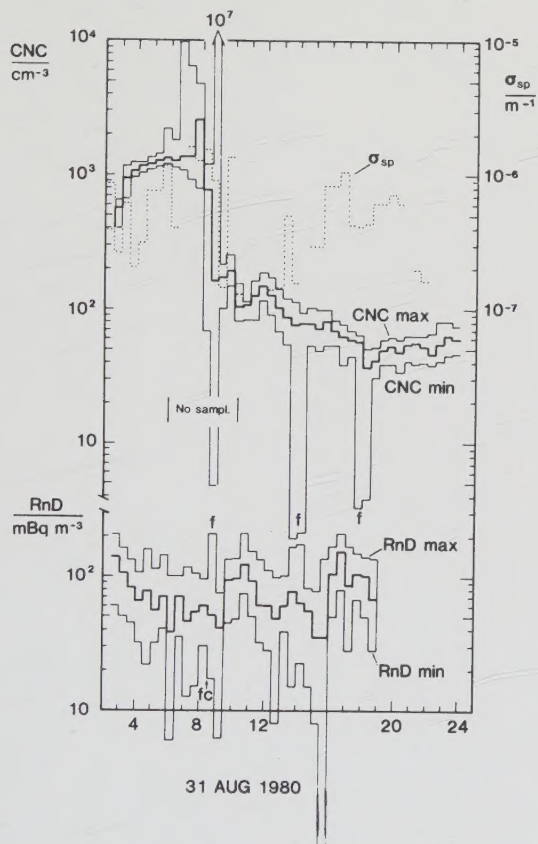


Figure 15.

